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International Congress of Pure and Applied Chemistry.

ORIGINAL COMMUNICATIONS

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OF APPLIED CHEMISTRY

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September 4 to 13, 1912

SECTION V*c*
FUELS AND ASPHALT

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THE CLASSIFICATION OF BITUMINOUS AND RESINOUS SUBSTANCES

By Herbert Abraham
Bound Brook, N. J.

The terms bitumen, asphalt, resin, tar, pitch, etc., are in common use, yet it is surprising to note how inadequately they are defined in most of the standard works and textbooks. This may be explained by the fact that these words originally had limited meanings, but in keeping pace with the progress of science they were extended in scope until they completely outgrew their original bounds.

It is probable that each of the expressions at first pertained to the aggregate properties characteristic of some typical substance closely associated with the processes of daily life. Thus, the generic term "resin" originated in the word "rosin," and it even now alludes to substances resembling ordinary rosin in appearance and physical properties. Similarly our comprehension of the generic term "wax" is based largely on the physical characteristics of the oldest known wax; namely, common beeswax.

From time to time, as new substances were discovered in nature, or produced in the arts, the meanings of these words were arbitrarily extended to include them. This resulted in a certain amount of over-lapping and consequent confusion. As the chemistry of these substances was investigated, this was in certain cases adopted as an additional means of differentiation. But we are still forced to rely principally upon the physical characteristics of these groups of materials, for even today comparatively little accurate information is available with respect to their chemical composition. It is probable, however, that in the future this will be adopted as a basis of nomenclature.

The complexity of these forms and existing confusion becomes especially apparent in attempting to define and limit the scope of claims in letters patent. We are not infrequently confronted with this problem in patent controversies, owing to the relatively large number of patents classified under the heading of "Plastic Compositions," and involving the use of asphalts, pitches and the like.

The object of this paper is to submit a list of definitions and a system of classification gradually evolved by the author while engaged in "patent causes." In this connection, it might be stated that several of these definitions (*e.g.*, resin, bitumen, resinous substance and bituminous substance), have already been accepted by the courts in this country. The author hopes, therefore, that at least some of his findings may be of value to the committee now engagedin fixing the nomemclature of bitumens.

The definitions which will follow have been based on one or more of the following items:

- *Origin.*
  - Natural:
    - Mineral,
    - Vegetable.
    - Animal.
  - Artificial:
    - Obtained from distillation processes,
    - Obtained fron oxidation processes,
    - Obtained from sulphurization processes.
  - Variable.
- *Composition.*
  - In such cases where this is known with certainty.
- *Solubility.*
  - In water,
  - In carbon disulphide, benzol, etc.
- *Physical Properties.*
  - Hardness:
    - Liquid,
    - Viscous,
    - Semi-solid,
    - Solid.
  - Color in Mass:
    - Light,
    - Dark.

CHART OF BITUMINOUS AND RESINOUS SUBSTANCES.

- **BITUMINOUS SUBSTANCES**
  - Natural
    - Bitumens
      - Gaseous
        - Natural Gas
        - Marsh Gas.
      - Liquid or Semi Liquid
        - Petroleums
          - Paraffin Base.
          - Asphaltic Base.
          - Mixed
        - Malthas ("Mineral Tars")
      - Solid
        - Mineral Waxes
          - Ozokerite (Ceresine)
          - Montan Wax.
          - Hatchettite.
        - Mineral Pitches
          - Asphalt (um)
            - Pure or Fairly Pure
            - Calcareous
            - Siliceous
            - Earthy
          - Asphaltites
            - Gilsonite
            - Glance Pitch
            - Manjak
            - Grahamite
    - Pyro Bitumens
      - Solids
        - Occur Fairly Pure
          - Derived from Asphalts
            - Elaterite (Wurtzelite)
            - Albertite
            - Impsonite
          - Derived from Vegetable Growths
            - Peat
            - Lignite
            - Bituminous Coal
            - Anthracite Coal.
        - Mineral Matter Predominates
          - Bituminous Shists
          - Bituminous Shales
  - Artificial
    - Animal Origin
      - From Bones
        - Bone Tar and Bone-tar Pitch
      - From Fats
        - Fatty-acid Pitches
          - Stearine Pitch ("Candle Tar")
          - Stearine-wool Pitch
          - Wool-fat Pitch ("Wool Pitch")
          - Palm-oil Pitch
          - Cottonseed-oil Pitch
          - Cotton-oil-foots Pitch
    - Vegetable Origin
      - From Vegetable Oils
        - Fatty-acid Pitches
      - From Saps of Coniferae
        - Resin Pitch
      - From Wood + Roots of Coniferae
        - Pine Tar and Pine-tar Pitch
      - From "Hard" Woods (e.g. Oak, Maple, Birch + Beech)
        - Wood Tar and Wood-tar Pitch
    - Mineral Origin
      - From Petroleum
        - Water-gas Tar and Water-gas-tar Pitch
        - Sludge Pitch
        - Petroleum Asphalt (Petroleum Pitch)
        - Blown (Oxidized) Petroleum Asphalt.
      - From Malthas.
        - Asphalt (um)
      - From Ozokerite (etc)
        - Paraffin etc
      - From Peat
        - Peat Tar and Peat-tar Pitch
      - From Lignite (Brown Coal)
        - Lignite Tar (Brown-coal Tar) and Lignite Pitch (Brown-coal-tar Pitch)
      - From Bituminous Coal
        - Coal Tar and Coal-tar Pitch
        - Coke-oven-Tar and Coke-oven-tar Pitch
        - Blast-furnace Tar and Blast-furnace-tar Pitch
        - Water-gas Tar and Water-gas-tar Pitch
        - Generator-gas Tar and Generator-gas-tar Pitch
      - From Elaterite
        - Elaterite Pitch
      - From Bituminous Shales
        - Shale Oil and Shale-oil Pitch
- **RESINOUS SUBSTANCES**
  - Natural
    - True Resins
      - Common Resins
        - Resin (Colophony)
        - Certain of the Copals (Recent origin)
        - Sandarac
        - Shellac
        - Anime
        - Damar.
        - Elemi
        - Mastic
      - Fossil Resins
        - Amber
        - Certain of the Copals
    - Oleo Resins
      - Common Turpentine
      - Canada Balsam.
      - Venice Turpentine
      - Burgundy Pitch
    - Gum Resins
      - Ammoniacum
      - Asafoetida
      - Bdellium
      - Gamboge.
      - Myrrh
    - Mineral Resins
      - Ozokerite, etc
      - Asphalt (um)
      - Asphaltites
  - Artificial
    - Animal Origin
      - From Bones
        - Bone-tar Pitch
      - From Fats
        - Fatty-acid Pitch
    - Vegetable Origin
      - From Vegetable Oils
        - Fatty-acid Pitch
        - Oxidized Oils
      - From Saps of Coniferae
        - Resin Pitch
      - From Wood and Roots of Coniferae
        - Certain Varieties of Pine Tar.
        - Pine-tar Pitch
      - From "Hard" Woods
        - Wood-tar Pitch
    - Mineral Origin
      - From Petroleum.
        - Water-gas-tar Pitch
        - Sludge Pitch
        - Petroleum Asphalts
        - Blown Petroleum Asphalts
      - From Maltha
        - Asphalt (um)
      - From Peat
        - Peat-tar Pitch
      - From Lignite
        - Lignite-tar Pitch
      - From Bituminous Coal
        - Coal-tar Pitch
        - Coke-oven-tar Pitch
        - Blast-furnace-tar Pitch
        - Water-gas-tar Pitch
        - Generator-gas-tar Pitch
      - From Elaterite
        - Elaterite Pitch
      - From Bituminous Shales
        - Shale-oil Pitch

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- Structure:
  - Amorphous.
- Fracture:
  - Conchoidal,
  - Hackly.
- Lustre:
  - Waxy,
  - Resinous.
- Feel:
  - Adherent,
  - Non-adherent,
  - Unctuous.
- Odor:
  - Tarry, etc.
- Behavior towards Heat:
  - Fusible,
  - Difficultly fusible,
  - Infusible.

DEFINITIONS

Bitumen: A naturally occurring[2] hydrocarbon complex,[7] often associated with a mineral matrix,[8] insoluble in water,[11] but largely soluble in carbon disulphide, benzol, etc.[13] Its color[24] and hardness[21] are variable.

Pyro Bitumen: A dark colored,[23] solid,[20] infusible,[35] naturally occurring[2] hydrocarbon complex,[7] often associated with a mineral matrix,[8] insoluble in water,[11] and relatively insoluble in carbon disulphide, benzol, etc.[14]

Bituminous Substance: A term applied to (*a*) Substances containing bitumens or pyro bitumens. (*b*) Substances resembling either the viscous or solid[17] bitumens, or pyro bitumens[18] in their physical properties; *i.e.*, having an amorphous structure[25] and dark color in mass.[23]

Maltha: A dark colored[23] and viscous,[15] naturally occurring[2] hydrocarbon complex,[7] insoluble in water,[11] but completely soluble in carbon disulphide, benzol, etc.[13]

Asphalt(um): A dark colored,[23] and more or less viscous to solid[17] hydrocarbon complex,[7] including: (*a*) The easily fusible bitumens[33] often associated with a mineral matrix,[8] not having a "waxy" lustre[27] or unctuous feel.[30] (*b*) Residuums obtained from the distillation,[3] oxidation,[4] sulphurization,[5] etc., of bitumens.

Asphaltite: A dark colored,[23] solid,[30] difficultly fusible,[34] naturally occurring[2] hydrocarbon complex,[7] insoluble in water,[11] but more or less completely soluble in carbon disulphide, benzol, etc.[13]

True Resin: A term applied to various vegetable principles occurring in, or obtained from the secretions or saps of certain plants and trees.[1] They are hard, fusible[33] and more or less brittle, non-adherent to slightly adherent[29] solids at ordinary temperatures[20] usually light colored in mass,[22] having an amorphous structure,[25] conchoidal fracture[26] and "resinous" lustre.[28] They are insoluble in water,[11] but more or less completely soluble in carbon disulphide, benzol, etc.[13] They are oxidation or polymerization products of the terpenes, and generally contain "resin" acids and esters.

Resinous Substance: A term applied to (*a*) Substances containing true resins. (*b*) Substances of variable origin[6] and composition[10] resembling true resins in their *physical* properties (with the exception of color), and solubility (*vide* True Resins).

Mineral Resin: A term applied to the solid bitumens (*vide* Bitumen).

Oleo Resin: The viscous to semi-solid[16] and usually light colored solutions of true resins in essential oils,[9] either obtained from, or constituting the saps of certain plants and trees.

Gum Resins: The semi-solid to solid [19] and usually light colored[22] emulsions or mixtures of true resins with various gums (carbo hydrates), constituting the sap of certain plants and trees, partly soluble in water.[12]

Tar: A dark colored,[23] bituminous substance, liquid or semi-liquid at room temperature, often possessing a characteristic "tarry" odor,[31] usually insoluble in water,[11] but miscible with carbon disulphide, benzol, etc.,[13] and which on distillation, oxidation, etc., forms a pitch. Its composition[10] and origin[6] are variable.

Pitch: A dark colored,[23] fusible[33] and more or less viscous to solid[17] bituminous or resinous substance, insoluble in water[11] but more or less completely soluble in carbon disulphide, benzol, etc.[13] Its composition[10] and origin[6] are variable.

Wax: A term applied to unctuous,[30] fusible[33] and more or less viscous to solid[17] substances, having a characteristic "waxy" lustre,[27] and which are insoluble in water,[11] but more or less soluble in carbon disulphide, benzol, etc.[13] They are extremely susceptible to changes in temperature.[32] Their composition,[10] origin[6] and color[24] are variable.

The terms "resinous substances" and "bituminous substances" overlap, as it were, for both of them include mineral waxes, asphaltum and asphaltites. This is made clear by the following table:

RELATION BETWEEN BITUMINOUS AND RESINOUS SUBSTANCES

Origin	*Bituminous Substances*			*Resinous Substances*	
Natural				True Resins	Vegetable Resins
				Oleo-Resins	
				Gum-Resins	
	Bitumens	Natural Gas			
		Petroleum			
		Mineral Wax	←→	Mineral Wax	Mineral Resins
		Asphaltum	←→	Asphaltum	
		Asphaltites	←→	Asphaltites	
	Pyro-Bitumens	Peat			
		Lignite			
		Bitum. Coal			
		Anthr. Coal			
		Metam. Asphalts			
Artificial	Tar				
	Pitches		←→	Pitches	
	Artificial Mixtures			Artificial Mixtures	
				Synthetic Products	
				Oxidized Oils	

Pyro bitumens, asphalts, tars, pitches and waxes admit being classified according to their origin as follows:

Pyro Bitumen: (*a*) Derived from the metamorphosis of vegetable growth; (*e.g.*, peat, lignite, bituminous coal and anthracite coal). (*b*) Derived from the metamorphosis of asphalts; (*e.g.*, elaterite or wurtzilite, albertite and impsonite).

The distinguishing features of the pyro bitumens derived from the metamorphosis of asphalts are as follows:

	Streak	Sp. Gr. At 77° F.	Melting Pt. (K. & S. Method)	Fixed Carbon	Fracture	Lustre	In Flame
Elaterite (Wurtzilite)	Light Brown	1.05-1.07	Decomposes	*5-25%*	—(*a*) Conchoidal — —(*b*) Hackly —	Bright Fairly Bright	Softens and burns. Softens and burns.
Albertite	Black	1.07-1.10	Decomposes	*25-50%*	—(*a*) Conchoidal —(*b*) Hackly	Bright Dull	Intumesces. Intumesces.
Impsonite	Black	1.10-1.20	Decomposes	*50-57%*	Hackly	Dull	Dcrepitates.

Asphaltum: (*a*) Natural — Resulting from a slow natural process of metamorphosis, known as natural asphalts ("mineral pitches").

1. Occurring in a fairly pure state.
2. Associated with a mineral matrix (*e.g.*, calcareous, siliceous or earthy).

(*b*) Artificial — Resulting from the distillation, oxidation, etc., of mineral oils; known as petroleum asphalt ("petroleum pitches").

1. Straight petroleum asphalt.
2. Cut back.
3. Blown (oxidized), petroleum asphalt.
4. Dubb's (sulphurized) petroleum asphalt.

Asphaltites: The distinguishing features of the individual asphaltites are as follows:

	Streak	Sp. Gr. At 77° F.	Melting Pt. (K. & S. Method)	Fixed Carbon	Fracture	Lustre	In Flame
Gilsonite (Uintaite)	Brown	1.06-1.10	250-350	*10-20%*	(*a*) Conchoidal (*b*) Hackly	Bright Fairly bright	Softens and flows. Softens and flows.
Glance Pitch	Brown to Black	1.10-1.25	250-350	*20-30%*	(*a*) Conchoidal (*b*) Hackly	Bright Fairly bright	Variable. Variable.
Manjak	Black	1.15-1.175	350-450	*30-40%*	(*a*) Conchoidal (*b*) Hackly	Bright Fairly bright to dull	Softens, splits and burns. Softens, splits and burns.
Grahamite	Black	1.175-1.20	450-550	*40-50%*	(*a*) Conchoidal (*b*) Hackly	Bright Fairly bright to dull	Decrepitates violently. Softens, splits and burns.

Tar: (*a*) Natural—(*e.g.*, mineral tars or maltha). (*b*) Artificial—The soft residues obtained.

1. From the distillation of petroleum (*e.g.*, petroleum tar or soft petroleum asphalt).

2. From the destructive distillation of organic substances and pyro bitumens; (*e.g.*, pine tar, wood tar, bone tar, coal tar, mineral tar (maltha), petroleum tar, etc.).

Pitch: (*a*) Natural — resulting from a slow natural process of metamorphosis known as "mineral pitches" or natural asphalts; *e.g.*, Trinidad pitch, glance pitch, gilsonite, etc. (*b*) Artificial —

1. Residues from the distillation, oxidation, etc., of mineral oils; *e.g.*, petroleum pitch, blown petroleum asphalt, sludge pitch, Dubb's asphalt, etc.

2. Residues from the distillation of tars; *e.g.*, coal-tar pitch, brown-coal-tar pitch, coke-oven-tar pitch, blast-furnace-tar pitch, water-gas-tar pitch, generator-gas-tar pitch, wood-tar pitch, pine-tar pitch, etc.

3. Residues from the distillation of fusible organic substances, the process having been terminated before the actual formation of coke; *e.g.*, resin pitch, stearin pitch (fatty acid pitches), etc.

4. Artificial mixtures complying with the above general definition of the term "pitch," regardless of their origin or composition (*e.g.*, insulating pitch, Brewer's pitch, roofing pitch, etc.).

Wax: (*a*) Natural —

1. Mineral (*e.g.*, ozokerite, ceresine, montan wax, hatchettite, etc.)

2. Vegetable (*e.g.*, Japan wax, carnauba wax, etc.)

3. Animal (*e.g.*, beeswax, spermaceti, etc.)

(*b*) Artificial —

1. From the distillation of paraffinaceous petroleum, ozokerite, etc.

2. From the destructive distillation of lignite (brown coal), paraffinaceous shales, etc.

The following chart shows the relation between bituminous substances, bitumens, pyro bitumens, maltha, asphaltum, asphaltite, tars and pitches; also between resinous substances, true resins, oleo resins, gum resins and mineral resins.

[1] Vegetable: These may be of recent formation or ancient formation. In the latter case, they are termed "fossil." It is usually conceded, however, that in either event they are of vegetable origin.

[2] Naturally Occurring: This excludes artificial products, as for example, certain tars and pitches, synthetic products, artificial mixtures, etc.

[3] Obtained from distillation of bitumens: This includes petroleum pitch, either "straight" or "cut back."

[4] Obtained from the "oxidation" of bitumens: This includes "blown" petroleum pitches.

[5] Obtained from the "sulphurization" of bitumens: This includes the so-called "Dubb's asphalt," obtained by heating petroleum residues with sulphur.

[6] Variable Origin: It may be of animal, vegetable or mineral origin and includes both natural and artificial products.

[7] Hydrocarbon Complex: Often contains oxygen, nitrogen or sulphur derivatives of the hydrocarbons. This excludes vegetable resins, vegetable and animal waxes, etc.

[8] Associated with a mineral matrix: This may be calcareous, siliceous, earthy, etc., which is usually present as an impurity.

[9] Solutions in essential oils: The essential oils consisting of terpenes.

[10] Variable Compositions: No exact chemical composition. Usually mixtures of closely related substances, composed of the elements carbon and hydrogen, together with varying amounts of oxygen, sulphur and nitrogen. May contain hydrocarbons, fats, fatty acids, resin acids, waxes, etc., etc., or mixtures of these.

[11] Insoluble in water: This excludes gum resins, carbo hydrates, etc.

[12] Partly soluble in water: The carbo hydrates ("gums") are the soluble constituents.

[13] Soluble in carbon disulphide, benzol, etc.: This does not take into consideration the mineral constituents present and excludes the pyro bitumens.

[14] Relatively insoluble in carbon disulphide, benzol, etc.: This likewise does not take into consideration the mineral constituents present and excludes the bitumens proper, tars and most pitches.

[15] Viscous: This excludes gaseous and solid bitumens.

[16] Viscous to semi-solid: This is governed by the amount of essential oil present.

[17] Viscous to solid: Some occur as sticky masses, others as hard and brittle solids at ordinary temperatures. Plasticity is an inherent or an acquired property. Some are plastic at ordinary temperatures, others become plastic, under the influence of heat or when combined with suitable fluxes. This excludes the gaseous and liquid bitumens, tars and oleo resins.

[18] Viscous to solid Bitumens and Pyro Bitumens: This includes the hard and relatively insoluble wood tars, pitches, etc.

[19] Semi-solid to solid: Usually contains small amounts of essential oils, which largely govern the consistency.

[20] Solid: This excludes gaseous, liquid and viscous substances.

[21] Variable Hardness: This includes gaseous (in the case of bitumens), also liquid, viscous and solid substances.

[22] Light Colored in Mass: This excludes the mineral resins.

[23] Dark Colored in Mass: Usually black. This excludes paraffin (ceresine), and vegetable resins.

[24] Variable Color in Mass: Color ranges from pure white (paraffin wax), to very dark, almost black (ozokerite and montan wax).

[25] Amorphous Structure: In distinction to crystalline.

[26] Conchoidal Fracture: But sometimes "hackly."

[27] Waxy Lustre: Mineral waxes have a waxy lustre, and are, therefore, included.

[28] Resinous Lustre: This together with the light color is characteristic of the true resins.

[29] Non-adherent to slightly adherent feel: In distinction to the unctuous feel of waxes.

[30] Unctuous Feel: This is characteristic of waxes.

[31] Tarry Odor: Most tars have a characteristic odor, but some mineral tars (*e.g.*, maltha, petroleum tars, etc.), are practically free from odor.

[32] Extremely susceptible to changes in temperature: Meaning that they pass *rapidly* from the solid to the liquid state as the temperature is raised (*i.e.*, have a high "susceptibility factor").

[33] Fusible: Soften on warming, melt *without* decomposition, decompose as the temperature is increased, and finally burn with a smoky flame, leaving a carbonaceous residue behind. This excludes the asphaltites, as these are difficultly fusible, and the pyrobitumens as these are infusible (but which, however, decompose and melt at higher temperatures).

[34] Difficultly Fusible: Intermediate between the natural asphalts and pyro bitumens.

[35] Infusible: This excludes the Bitumens Proper, Vegetable Resins, Tars, Pitches, etc.

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METHODS FOR THE DETERMINATION OF WATER IN PETROLEUM AND ITS PRODUCTS

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Methods employed for the determination of water in petroleum and in its products may be classified as follows:

1. Loss in weight on heating.
2. Diluting with a solvent and settling by gravity.
3. Diluting with a solvent and separating with a centrifuge.
4. Treating with a calcium carbide.
5. Treating with sodium.
6. Use of a color comparitor tube.
7. Treating with normal acids.
8. Electrical treatment.
9. Distilling with a nonmiscible liquid.
10. Directly distilling off the water.

1. Loss in Weight on Heating

Conradson determines the water content of heavy oils by the loss in weight on heating. However, he recognizes the fact that when a heavy oil is heated to a temperature of 100° C. some oil will be volatilized and lost.

Wright heats to 105° C., Hurst to 220° F., and Archbutt and Deeley to 105-110° C. Davis saturates a tarred filter paper and heats to 110°C., Patrick heats the oil until slight decomposition takes place, and assumes the loss in weight to be a loss of water.

Gavaloski has improved the evaporation method by using duplicate samples in tared flasks. He heats one flask, opened in the usual manner and attaches to the other a sulphuric-acid drying tube, which, he states, absorbs the water and allows the oil vapors to pass through unabsorbed. The difference in loss of weight he assumes to be a water loss. Lowenstein dilutes with 95 per cent alcohol and dries repeatedly on a water bath and then at a temperature of 105° C. to constant weight.

The evaporation method varied, as described above, is approximate and applicable to heavy oils and greases only. Its accuracy even with heavy greases is questionable.

2. DILUTING WITH A SOLVENT AND SETTLING BY GRAVITY

Curtus and Thompkins dilute 200 cc. of the oil with 600 cc. of 70° Baumé gasoline, allow the liquid to settle in a graduated glass tube, and read the volume of water settling to the bottom of the tube. Sussanow in using this method employs benzine instead of gasoline as a solvent.

This method is only approximate. Its accuracy is modified by the quantity and quality of the diluent used, by the amount of mineral matter in the oil, by the character of insoluble portions of the oil itself, such as asphaltine, and by the length of time allowed for settling.

3. DILUTING WITH A SOLVENT AND SEPARATING WITH A ENTRIFUGE

Charitschkow, Wieloezynski, Rosenthal, and O'Neill have greatly improved the method. After diluting the oil with an equal volume of benzine they centrifuge vigorously until the separated layer of water does not appear to increase in volume. However, as water is somewhat soluble in any diluent used and also in oils, a portion of the water content will fail to appear, so that consequently the method in which a diluent is used cannot be considered accurate. It is advisable first to agitate the diluent vigorously with water and then centrifuge in order to saturate it with water before using.

Grosschuff states that 100 grams of benzine at 3° C. will dissolve 0.03 grams of water, and 3.37 grams at 77° C., whereas petroleum products (density 0.792) will dissolve from 0.012 grams at 2° C. to 9.7 grams at 94° C.

4. TREATING WITH CALCIUM CARBIDE

Dupre, Danne, O. Masson and I. Masson agitate samples of the various materials containing water under examination with an excess of calcium carbide, measure the volume of acetylene evolved, and calculate the content of water.

Wielezynski, Roberts and Fraser maintain that this method may be applied successfully to petroleum. The method may be carried out conveniently as follows:

To the stopper of a 250 cc. separatory funnel *a* is fitted a second 100 cc. separatory funnel *b*, which in turn is fitted with a small glass stop cock *c*; 17.24 grams of the sample is run into *a*, the lower cock of *b* is closed and *b* is connected to *a*. About 10 grams of powdered calcium carbide is run into *b* and cock *c* inserted. Cock *b* is cautiously opened, the dropping of any carbide into *a* being avoided, and the whole is immersed in a water bath maintained at any convenient temperature *t*. Immersion is made up to cock *c*, which is open to the atmosphere. When the temperature *t* has been reached *c* is closed, the apparatus is removed from the bath and the carbide is shaken into *a*. The sample and carbide are then shaken vigorously, and, if necessary, are cooled several times under cold water. After the reaction is completed, the whole is again brought to temperature *t* by immersion in the water bath, union is made with *c* and a gas buret, and the increase in volume due to the generation of acetylene is measured. One gram of water generates approximately 580 cc. of acetylene at 0° C. and 760 mm. pressure.

It should be noted that acetylene is soluble in oils. One cc. of kerosene (specific gravity 0.7950) dissolves approximately 1.375 cc. of acetylene, and 1 cc. of spindle oil of specific gravity 0.8667 will dissolve approximately 0.95 cc. of acetylene, that is an average of 1.10 cc. of acetylene per 1 cc. of oil. This absorption is not a constant but will vary with different oils. This variation will introduce an error of approximately 110 cc. per 100 cc. of oil used; or 17.24 grams of a sample will absorb 18.96 cc. of acetylene, introducing an error of approximately 18.96 in 580 cc., that is 3.2 per cent in the water content. For this error, approximate correction can, of course, be made. If the sample is too viscous to mix readily with the carbide it may be diluted with 50 cc. of an oil of low vapor tension such as kerosene, and correction may be made for the acetylene absorbed therein. For example, 55 cc. of acetylene absorbed in 50 cc. of the diluent plus 18.96 cc. absorbed in the 17.24 cc. of sample will give an approximate correction of 74 cc., which is to be added to the buret reading. It must also be noted that a portion of the water is held by the calcium hydroxide formed in the action of the water on the carbide.

This method is easily carried out, is approximately correct, and may be used for many classes of materials containing water.

5. Treating with Sodium

Græfe estimates the water content of an oil by agitating with finely cut metallic sodium.

This may be carried out as follows:

For each gram of water in the sample of oil, about two grams of finely cut sodium is used in the same apparatus and in exactly the same manner as described in Method 4.

This will react in the following manner:

$$Na_2 \quad 2\,H_2O \quad 2\,NaOH \quad H_2$$

or 1 g. of water generates $\frac{2}{36}$ g. of hydrogen.

1000 cc. of hydrogen weigh 0.09004 g. at 0° C., and 760 mm. pressure or 1 g. of hydrogen is 11,106.17 cc.

One g. of water generates $\frac{2}{36}$ (11,106.17) cc., or 617.0095 cc. of hydrogen, or 1 cc. of hydrogen represents .001604 g. of water in the sample used.

This gives much more accurate results than when calcium carbide is used, as hydrogen is not appreciably soluble in petroleum products.

6. Use of a Color Comparitor Tube

Martin compares the sample under consideration in a color comparitor tube with standard mixtures of oil and water. This method of determination of water content is roughly approximate and applicable only when the water is present in visible suspension.

7. TREATING WITH NORMAL ACIDS

Nettel mixes 100 cc. of the sample with 100 cc. of petroleum spirit, adds 50 cc. of tenth-normal hydrochloric acid, agitates five minutes, allows the mixture to settle at 65° C. for half an hour, and titrates an aliquot part of the separated acid-water layer with normal alkalies. He concludes that the loss in acidity of the standard solution is due to the absorption of acid by the water dissolved in the oil, basing his conclusions on the assumption, first, that water in petroleums is neither acid nor alkaline; second, that the added acid solution mixes thoroughly with the water in the oil; and third, that crude oil itself has no action on the acid.

Crude oil often contains natural mineral waters which may be either acid or alkaline, and refined oils may often contain some of the wash alkalies. The presence of such mineral waters or wash alkalies would prevent accurate results from the use of the method described above.

8. ELECTRICAL TREATMENT

A convenient method to reduce the water content of an emulsified oil to approximately 0.5 per cent, is as developed by Cottrell and Speed. The method is admirably adapted for reducing the water content on a large scale. A convenient laboratory apparatus may be made as follows:

A glass tube about 5 cm. in diameter and 30 cm. long, and open at the top, is drawn out at its lower end and fitted with a glass cock. On the inner surface of the tube is fitted a copper gauze cylinder extending from the top to within 5 cm. of the bottom. Between the gauze and the tube is placed a single wrapping of wet cheese cloth or very thin muslin. The tube, held in a vertical position, has centered within it a heavy copper wire or rod which also extends from the top to within 5 cm. of the bottom. The copper wire and the copper-gauze cylinder are connected to the high-potential (2200 volts) terminals of a step up transformer.

In use the tube is filled with the emulsified oil to be examined, and the circuit is closed. The high-tension current breaks down the emulsion and allows the water to trickle down the cheese cloth and settle into the lower end of the tube, where it can be drawn off. With oils containing considerable water, the current must be broken at short intervals to prevent over-heating of the oil.

This process will readily reduce the water content of an emulsion, with little heating, to as low as 0.5 per cent in half an hour, and is admirably adapted to reduce the water content of emulsions when heating is objectionable, but quantitatively can not be compared with methods 2, 3, 4, 5, 9 and 10.

9. DISTILLING WITH A NONMISCIBLE LIQUID

Dean estimates the moisture in creosote as a result of distillation with a water-saturated xylene and of measurement of the water that separates in the distillate. Holde and Marcusson use the same method for water in greases, whereas Marcusson uses toluene, as well as xylene.

Testoni employs turpentine in a like manner for the determination of the water content of molasses; Prutzman uses a mixture of benzine and toluene for this purpose. Sadtler recommends the use of benzine boiling at a temperature of 300 to 450 F.

A convenient method is as follows:

One hundred grams of the sample is mixed in a distilling flask with 200 grams of toluene (or benzine or xylene) and distilled into a graduate in the usual manner until the distillate comes over perfectly clear. The distillate is allowed to settle by gravity, or is separated by a centrifuge, and the volume of water is read off directly, or the water may be withdrawn with a micropipette and weighed. As water is somewhat soluble in all of the three hydrocarbons mentioned, the method is not accurate but is convenient when approximate results only are desired.

10. DIRECTLY DISTILLING OFF THE WATER

Bell, Senger and Allen distill the sample directly and measure or weigh the quantity of water settling out in the distillate.

The water content may be accurately and conveniently determined during the course of an ordinary distillation in the following manner:

Two hundred grams of the sample is weighed into a one-quarter-liter distilling flask and the distillation carried out in the ordinary manner at the rate of one drop of distillate per second. The distillation can be performed most accurately in an electric still. At temperature between 100 and 150° C., the water distills over and can be removed from the receivers by means of a micropipette and weighed. Usually a few drops of water adhere to the condenser and fail to run into the receivers; in this event a small pellet of absorbent cotton, moistened with water, squeezed as dry as possible, and weighed, is fastened to a wire and run up into the condenser to remove these last traces of water. The amount of the increased weight of the cotton pellet, reckoned as water, is added to the weight of the water in the receivers.

With an oil containing considerable water, it is advisable to cause a slow current of a dry inert gas, such as carbonic acid, to bubble through the oil in the distilling flask to carry off the vapors of oil and water as soon as formed. The gas current will reduce bumping and overheating of the oil to a minimum. The condenser must also be kept well cooled throughout the distillation. This method, with the quantity of sample and the apparatus above mentioned, is accurate to less than 0.03 per cent.

Conclusions

Method 1 is approximate and applicable to heavy oils and greases only.

Method 2 is approximate and applicable to thin oils. A diluent is to be avoided.

Method 3 is like Method 2, but is more rapid.

Method 4 is convenient and, with petroleums, accurate to approximately 3 per cent of the water percentage.

Method 5 is convenient and accurate.

Methods 6 and 7 are approximate only.

Method 8 is successful in breaking up an emulsion on a commercial scale or in reducing the water content of an oil to such a condition that it can be successfully treated in some other manner.

Method 9 is accurate to approximately .033 grams of water per 100 cc. of benzine and oil in the distillate.

Method 10 is convenient and can be used simultaneously with a distillation. It is accurate to about .003 grams of water in the distillate if the water be cooled to about 2 C.

1. DRY FUSION WITH ALKALIES AND SUBSEQUENT OXIDATION WITH BROMINE

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Heath and Sadtler, in determining the total sulphur in a given fuel, fuse the sample directly with dry sodium carbonate and magnesium oxide and eventually complete the oxidation with bromine and precipitate with barium chloride in the usual manner the sulphuric acid formed.

Handy fuses the sample with either sodium or potassium carbonate.

Hundeshangen and Herting fuse with a mixture of sodium and potassium carbonates and magnesium oxide. Hundeshagen fuses with magnesium oxide and potassium hydrate. Garrett and Lomax prefer to fuse with calcium oxide and sodium carbonate because they maintain that calcium oxide free from sulphur is more easily obtained.

A usual method of procedure is as follows:

In a platinum crucible of 10 cc. capacity 0.7 to 1.5 grams of the sample is intimately mixed with 3 to 4 grams of a fusion mixture, consisting of 4 grams of magnesium oxide, 1 gram of sodium carbonate, and 1 gram of potassium carbonate. The crucible is then filled with magnesium oxide. Over this is inverted a somewhat larger platinum crucible. The two crucibles are then inverted, the larger is nearly filled with the fusion mixture, and is loosely covered with the crucible lid. The crucible is tilted about 15° in a porcelain triangle and heated very gradually, avoiding spurting, over a sulphur-free flame till the crucible is red hot. This heating requires about 20 minutes. The crucibles are then cooled, boiled in 100 cc. of water containing 15 cc. of bromine water, acidified with hydrochloric acid, filtered, boiled free from carbonic acid, and treated with a slight excess of boiling barium (chloride solution) in the usual manner.

2. DRY FUSION WITH A MIXTURE OF ALKALIES AND OXIDIZING AGENTS

Blair, Peckham, Lidow, and Koch and Upson fuse the sample with sodium carbonate and potassium nitrate, or with ammonium nitrate and sodium carbonate.

Schreiber fuses with sodium hydroxide, sodium nitrate, and magnesium nitrate.

Langmuir fuses with potassium carbonate and potassium nitrate. Aufrecht fuses with acid sodium carbonate and ammonium nitrate. Eschka fuses with magnesium oxide and sodium carbonate. He sometimes adds ammonium nitrate. Charitschkoff treats with potassium bichromate and fuming hydrochloric acid and completes the oxidation with bromine.

In using this method, one may proceed as under Method 1, using, however, as a fusion mixture 4 grams of magnesium oxide, 1 gram of sodium carbonate, and 1 gram of potassium nitrate, thus avoiding the use of bromine. It is necessary in this case after fusion to evaporate to dryness repeatedly with concentrated hydrochloric acid to expel the last traces of nitric acid before precipitating with the barium chloride. It should be noted that this fusion should not be made in platinum, because if more than .02 grams of nitrates is used in the fusion, they will attack the platinum.

The expulsion of nitric acid is troublesome, so that this method is not as satisfactory as Method 1.

Furman and Iles fuse the sample with potassium hydrate and treat with bromine.

Dubois fuses with sodium carbonate, manganese dioxide, and potassium permanganate.

Schillbach fuses with barium peroxide.

Osborne, Parr, Wheeler, and Berolzheimer, Konek and St. Warunis fuse with sodium peroxide.

Method 2 has its adherents, but there seems to be little to say for or against it as compared with Method 1, except that it is less convenient.

3. TREATING WITH WET ALKALIES AND OXIDIZING AGENTS

Lecocq and Vandervoort treat the sample with a solution of hydrogen peroxide.

Gill and Grindley treat with nitric acid and potassium nitrate.

Goetzl concentrates the sample to a small bulk with fuming nitric acid, then fuses with sodium carbonate and potassium nitrate.

Calvert boils 20 hours with nitric acid, Marchlewski extracts with carbon-bisulphide, concentrates, and oxidizes with nitric acid and potassium perchlorate.

Schulz boils with a 30 per cent solution of sodium hydroxide containing zinc and lead acetate, filters the lead sulphide formed, and fuses with sodium and potassium carbonates.

Method 3, with the modifications mentioned, also has its adherents, but like Method 2 seems to be less convenient than Method 1.

All of the above-mentioned fusion processes must be carried out most carefully under a slow heating, and all require a considerable time for their completion. Though sufficiently accurate for such dry materials as coal and coke, the readiness with which oils volatilize and burn prohibits their use in oil analysis.

4. OXIDATION WITH FUMING NITRIC ACID AT HIGH PRESSURES

Carius seals a given fuel sample with fuming nitric acid in a glass tube and heats under pressure to complete the oxidization.

Anelli claims that by introducing barium nitrate into the Carius tube more accurate results are obtained.

Holand suggests the introduction of potassium perchlorate into the Carius tube.

Method 4, though accurate, on account of the tendency of the tube to explode under heating, is difficult to make and is not universally satisfactory.

5. BURNING IN PURE OXYGEN AT ATMOSPHERIC PRESSURE

Hempel burns the given fuel sample in oxygen in a large glass bottle, and completes the oxidization with bromine.

Gräfe treats in a like manner, but completes the oxidization with sodium peroxide solution.

The method may be carried out as follows:

Into the stopper of a 10-liter glass bottle is fitted a dropping funnel of 100 cc. capacity filled with a five per cent solution of hydrochloric acid containing a few drops of bromine. Two platinum wires to carry the ignition current are passed through the stopper. One wire supports a small crucible containing the sample. A microplatinum wire or a pure iron wire (No. 36 B. & S. gage) short circuits the two wires and dips into the fuel, which, when the current is used, acts as an incinerator to ignite the sample. The bottle is filled with oxygen and the stopper tied firmly in place. On ignition the sample burns. The bottle is then cooled by immersing in cold water, the contents of the dropping funnel are run into the bottle to complete the oxidation, the contents of the bottle are washed into a beaker, and the sulphuric acid is precipitated as under Method 1 or Method 8.

The manipulation of the above-described apparatus is difficult, the oxidations are not dependable, and the method has not received general application.

6. Burning in a Stream of Pure Oxygen

The elemental organic combustion train is adaptable to the determination of sulphur. The sulphur-determination method involved in the use of this train, as modified by Mabery, Barlow and Tolleus, Brugelmann, Bay, Warren, Sauer, and Mixter, is practicable and accurate though requiring a considerable degree of care.

Dammer has further improved this method by introducing in the train platinized asbestos as an oxygen carrier.

Dennstedt uses platinized quartz, whereas Brunck mixes the sample with cobalt oxide. These catalizers have greatly improved the general usefulness of the method.

Dennstedt proceeds as follows:

The ordinary elemental organic combustion train with a tube of hard glass is employed. The train is fitted with the usual washing bottle at the entrance of the furnace and with absorbing potash bulbs at its exit. At the entrance, however, the combustion tube is fitted with an inner tube *b* holding the boat, which communicates directly with the oxygen supply independently of the combustion tube itself. This arrangement allows the operator to pass oxygen either directly over the sample, thereby causing rapid combustion, or he may pass oxygen into the outer tube, from whence it may unite with the sample after volatilization.

By this arrangement the operator can accelerate or retard the combustion at will, a necessity in working with volatile oils. The combustion tube, for about two inches of its length, is filled with platinized quartz or asbestos instead of being filled for 10 inches with copper oxide, as in the older furnace. The quartz or asbestos acts as an oxygen carrier to complete the combustion. The sulphuric-acid fumes are absorbed directly in potash bulbs, or, if a carbon-hydrogen determination is made simultaneously, lead superoxide in two long boats is placed in the end of the tube. The lead superoxide absorbs the sulphuric-acid fumes, which later may be leached with a 25 per cent solution of sodium carbonate, and the sulphuric acid may be precipitated therefrom with barium chloride in the usual manner described under Methods 1 and 8.

This combustion method must be carried out with considerable care, however, to avoid too rapid a volatilization of the oil, with consequent incomplete oxidation. It requires the constant vigilance of the operator and a considerable time for completion, but gives admirable results.

7. BURNING IN A LAMP IN ATMOSPHERIC OXYGEN

A method applicable for the determination of sulphur in burning oils is that of burning a given oil in a small lamp and determining the sulphur in the products of combustion.

Fischer, Engler, Allen, Martwin-Riga use a process that embodies the absorption of the products of combustion, in a dilute potassium or sodium hydroxide solution, completion of the oxidization with bromine, and precipitation of the sulphuric acid thus formed with barium chloride, in the usual manner.

Housler, Kissling, Friedlander complete the oxidation of the products of combustion with potassium permanganate.

Magruder and Burton absorb the products of combustion in normal sulphuric acid.

The process of Magruder *a* and Burton *b* embraces the absorption of the products of combustion in normal sulphuric acid and titration with normal sodium carbonate, using 00 tropæoline as an indicator.

For all burning oils, this method is practicable and accurate, but as Conradson states, a considerable portion of the sulphur in the form of sulphates and sulphonates remain absorbed in the wick of the lamp. This must be determined by a fusion method, correction being made therefor.

8. BURNING IN A CALORIMETRIC BOMB UNDER 30 TO 40 ATMOSPHERES PRESSURE OF PURE OXYGEN

The method found most practicable and accurate for all oils, and the one used almost without exception in the petroleum laboratory of the Bureau of Mines, is the calorimetric-bomb method. Allen, Lord, and Lohmann proceed as follows:

0.7 to 1.0 grams of the sample is burned in a Berthelot calorimetric combustion bomb containing 10 cc. of water and 30 atmospheres of oxygen. (A lower pressure of oxygen will sometimes give inaccurate results.) If the sample contains more than 3.0 per cent sulphur the bomb is allowed to stand in its water bath for 15 minutes before opening. If the sulphur content is as high as 5.0 per cent, 40 atmospheres of oxygen is used. With these high oxygen pressures repeated trials have failed to show even traces of carbon monoxide or sulphur dioxide when a Berthelot bomb of 500 cc. to 600 cc. capacity is used. With a smaller bomb, such as the Peters or Kroker, of about 175 cc. capacity, incomplete combustion may result if too large a sample be taken, due to an insufficiency of oxygen.

A. C. Fieldner, assistant chemist of the bureau, in a conversation with the authors, stated that "a mixture of pyrite and coal was made, the mixture containing 25 per cent sulphur. One gram of this mixture was burned in a bomb under a pressure of 18 atmospheres. Upon opening the bomb an odor of SO_2 was noticeable. Burned at 30 atmospheres the sulphur was 0.2 per cent lower than when determined by the Eschka method." The combustion would have been complete if a pressure of 40 atmospheres of oxygen had been used. After cooling (15 minutes' standing usually suffices) the bomb is opened and its contents washed into a beaker. If the bomb has a lead washer 5 cc. of a saturated solution of sodium carbonate is added, the contents are heated to the boiling point, boiled for 10 minutes, and are then filtered. This operation is necessary to decompose the lead sulphate that might have shipped off from the washer. The united washings are then filtered, acidified with hydrochloric acid, boiled to expel all carbonic acid, and the sulphuric-acid content is determined in the usual way with barium chloride.

Gravimetric determination is preferred to volumetric because the nitrogen contained in the air originally in the bomb is oxidized in part to nitroacids, which cause a small error if the volumetric determination alone is used. The sulphur content of any combustible material from light gasoline (weighed in a tared gelatin capsule) to solid bitumens and cokes can be readily determined by this method.

The method of burning in a bomb is accurate, practicable, and rapid and is recommended over all of the other methods described above. It can be made simultaneously with the calorimetric determination if the latter be desired.

CONCLUSIONS

Methods 1, 2 and 3, though successful with such slow-burning materials as coals and cokes, are not dependable with oils.

Method 4 is accurate but because of frequent explosions is unsatisfactory.

Method 5 is not accurate because at atmospheric pressure the combustion is not always complete.

Method 6 is accurate but is tedious and must be under the constant observation of the operator.

Method 7 is applicable to burning oils but not to gaseous oils or to heavy residues.

Method 8 is applicable to all oils and combustible materials, and is accurate, rapid, and dependable.

THE TECHNICAL PROBLEMS OF COAL PREPARATION

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The problems involved in the preparation of coal begin in the mine itself and before the coal is even cut, and end with the loading of the prepared product into transportation cars ready for market. For the solution of these problems the engineer must bring to his aid geology, petrology, mineralogy, chemistry, hydrostatics, pneumatics and mechanics. These sciences have, after much study and a long-continued experimentation with every variety of coal, divulged the limits within which their respective groups of natural laws can be used for the accomplishment of the end sought. Any practice outside of these definite limits is sure to lead to disaster.

Having these guiding principles at hand, the continuity, uniformity, and structural nature of the beds of coal are determined, from which the system of mining and cutting is formulated; the physical peculiarities of both the coal and the refuse are carefully studied and the methods of handling and preparation are definitely selected; the degree of purity possible in the preparation, the possible percentage of recovery, the economy of the process adopted, and the market value of the prepared product, are also determined.

In the mine it is of great importance to first determine the recoverable tonnage in the coal beds, and from this data to determine the daily capacity of the proposed preparation plant.

The method of mining the several seams should be governed chiefly by the manner in which the refuse is associated with the coal. The breaking of the slates and rock into small pieces, in the cutting or blasting of the coal, must be avoided as much as possible for the reason that the larger the pieces of refuse and the smaller the quantity of it that is finely broken the simpler and less expensive the preparation of the coal for market will be. Wherever a shaly roof clod is free to fall as soon as the coal is removed, every economic means should be resorted to to prevent it from mixing with the coal.

But it is also of very great importance in the preparation problem to determine what alterations, if any, have taken place in the coal beds from folds in the strata. These folds, wherever found, have altered to a greater or less degree the fracture of the coal. In many cases they have crushed the coal almost to a powder, particularly where they occur in the anthracite and semi-anthracite fields. In others, the folding has caused a shale-like fracture of both the coal and slate. Upon the presence of the flat fractured material, in considerable quantity, chiefly depends the selection of the preparation scheme. There are some other conditions, however, which may materially modify that selection, namely, the manner in which the refuse is associated or combined with the coal. It may be associated in a very simple way, consisting of bands of slate or rock a half inch or more in thickness, which separate from the coal, in the process of mining, by distinct dividing planes, or it may be combined with the coal without any partings, and can only be separated from it by hand tools or by repeated crushing in the preparation plant.

As to the transportation of the material from the working places in the mine to the preparation plant, it should be done in closed-end cars. The method of handling closed-end cars at the dump is simple enough after full provision has been made for the system. The saving of coal filtered through the doors along the gangways and haulage tracks, the saving in labor necessary to periodically clean up these tracks, the avoidance of accidents from the doors springing open, the saving in repairs to the mine-cars because of the absence of the car doors and the safer condition against dust explosions, would pay a dividend over any advantages that may exist in favor of the open-end or door-car.

The preparation of coal, whether anthracite, bituminous or lignite, or any of the grades between or beyond them, must make use of the same natural laws, namely, those governing the difference in specific gravity and those governing the difference in the angle of repose. One of these groups may be perfectly efficient in preparing one coal having certain physical characteristics, while for another coal the other group would do perfect work, and for the almost infinite mergings of the two typical classes of coal a series of combinations and modifications of the two principal methods would have to be employed.

In the anthracite fields of Pennsylvania, the preparation of coal has been carried to the highest degree of efficiency and economy known to the world, yet the problem in all its details is not yet satisfactorily solved.

In the earliest days of anthracite mining in Pennsylvania only pure lumps of coal were shipped, and to obtain these only the purest beds of the series or the purest benches of a single bed were mined. The fine coal which would pass through a mesh of about two inches was sent out to the waste-bank. No attempt was made to mine the entire bed, or to systematically prepare the coal either by sizing or by removing the impurities.

The governing factors in the preparation of any coal are, to send the coal to the consumer in a condition suited to his requirements, and at a cost that leaves a margin of profit to the operator. The consumer's requirements are presumably based on the economic results that he desires to obtain, whether they pertain to steam generation, gas production, or metallurgical processes. Therefore, to meet his requirements, — which set the standards for sizes and purity, — the producer must search out ways and means, within economic limits, to prepare his coal. The economic end of this problem is not an easy one, and consequently because of the extended and intricate nature of the preparation requirements for some complicated deposits of coal, added to a difficult mining proposition, there is but little profit left to the operator; therefore, it is easy to understand why one operation will not pay a profit, while another with a different deposit pays handsomely. Each must prepare its coal according to the same standards for size and purity, no matter what it may cost, and receive for its prepared product the same market value.

In the specific gravity process of preparation, typified in the jig, many perplexing problems have arisen and many annoying results are always present, depending for their degree of seriousness upon the physical nature of the material being treated.

Any material in order to be treated in the most efficient manner by any process, must first be sized. The most efficient method of sizing coal as it comes from the mine, — and that sizing must have a very limited range in variation, — is by a series of shaking screens. The revolving screen is a thing of the past, because of its inefficiency, its destructiveness to the coal by chipping, and its reversal of the natural order of sizing. The mesh of these shaking screens is circular, and it is self-evident that this circular mesh will allow, but in a less degree than the old square mesh, all conceivable shapes of material to drop through it, provided that each individual piece has for the maximum dimension of its minimum cross section a measurement just a little less than the diameter of the mesh. The result is that the material as sized contains cubes, pieces that are triangular, round, oblong, rectangular, and square, having thicknesses that are very small compared with their other dimensions, and an infinite gradation of sectional areas between each of these typical shapes and that of the cube. The specific gravity of the slate and rock ranges from 1.90 to 2.50, while the specific gravity of the coal averages about 1.50. It is a very easy matter for any one to demonstrate by a few simple calculations, using the specific gravities of the coal and refuse just given and applying them to the various shapes of pieces given, that a cube of pure coal will tend to pass out of the jig with the refuse, while the thin flat pieces of slate of the lighter specific gravity are sure to float over with the coal. The reason for this is that the ratio of the area of surface of the cube of coal, exposed to the impulses of the water in the jig, to its weight, is greater than the ratio between the area of surface exposed of a thin light piece of slate and its weight. These very inefficiencies are experienced wherever this class of variformed material is treated by the jig. The inherent conditions make the failure of the jig a natural consequence, and confirm the conclusion from tests that no jig can be made to separate efficiently a material containing a variety of forms, particularly cubical pieces of coal and flat pieces of light slate. Much time and money have been spent in a fruitless endeavor to meet the condition by introducing some new and untried form of jig, with the same inefficient results. A thorough grasp of the technical features of this problem would at once show clearly the limits of the possibilities of coal preparation by this process alone.

The attrition in the jig caused by its rapid pulsations amounts to a serious waste of coal, ranging from 2 per cent for the very hardest coal to 21½ per cent for the softest anthracite, or at the present market value of prepared sizes to from 8 cents per ton to 89 cents per ton. The average loss sustained is estimated to be between three and five per cent. The total loss per year by chippings alone on coal treated by the jig and separating machines causing impact in the anthracite field of Pennsylvania is estimated to be more than three million dollars ($3,000,000).

Of the other mechanical devices for removing impurities from coal there are three that are extensively used, each employing the difference in the angle of repose between slate and coal as the basic principle of its operation.

The first is so constructed that a broad stream of material, about 5 feet wide and spread out in such manner that the pieces in their forward movement will not touch each other, is intermittently fed upon a short adjustable incline floor, at the lower edge of which is a narrow section of slate or other gritty material. Immediately below the section of slate is a narrow opening of several inches and beyond this is a continuation of the inclined floor. The action is that the refuse passing over the slate section of the floor is retarded and drops through the narrow opening, while the coal because of its less angle of repose continues over the slate section with only slightly retarded velocity and jumps across the narrow opening and upon the continuation of the inclined floor below. The serious objection to this type of machine is the great loss due to the chipping of coal as it impinges against the upper edge of the continued floor below the opening. The loss is reported to be greater than that in the jig.

The second consists of a spiral floor descending and inclined inwardly toward its vertical axis. It depends for its operation upon the inward inclination of the floor toward the vertical axis tending to draw the slate in its retarded movement toward the axis, while the coal in its accelerated movement down the spiral incline is thrown from the floor by centrifugal force and is caught in a specially provided runway paralleling the spiral floor and spaced a little distance from it. This separating device is also very destructive to the coal as each and every piece as it leaves the spiral floor impinges against the outside of the runway. In quantity the loss is reported to be fully equal to that in the jig.

The third device consists of a moving floor inclined forwardly and transversely. The motion of the floor is upward on the transverse inclination. Both inclinations are adjustable. The material travels across the floor on its longitudinal inclination. The upwardly moving floor removes the impurities laterally from the forwardly moving stream. The coal slides upon the moving floor and across it without any impact. The loss sustained in this separator is less than one-half of one per cent. This machine is the only one that does not create material loss by attrition or chipping.

The tendency in the practice of today is to use as extensively as possible the machines that do not create loss, therewith turning to the pocket ready for market from 60 to 80 per cent of the pure coal coming upon the machine, and without appreciable loss from chippings. This operation removes all of the cubical coal which gives unsatisfactory results in the jig. A second operation supplements it, retreating the tailings of the first, which removes the flat slate and heavy rock. This operation is again supplemented by a third, jigging, which removes the remaining slate in the balance of the coal. This three-stage process has the advantages of great elasticity and controllability.

The most important of all features in the entire process of anthracite preparation is to so handle the coal in transportation, screening, separation, conveying and loading as to retain the pieces of coal whether coming directly from the mine or from the crushing-rolls in their initial form without any loss in weight from abrasion or chipping, if at all possible to do so. The crushing-rolls must be of the type that will yield the very highest percentage of the prepared sizes, for in these sizes the values lie. These sizes are egg, through 3 1-4 and over 2 5-16 inch round mesh; stove, through 2 5-16 and over 1 5-8 inch mesh; and chestnut, through 1 5-8 and over 15-16 inch mesh.

The foregoing reference to the losses sustained in the process of jigging and in certain types of the frictional machines accentuates the importance of this feature of the process.

Great attention should be given to the avoidance of breakage in the transportation of the coal from one stage of the process of preparation to the next and finally its delivery into the deep pockets holding as much as 100 to 200 tons for each size. One of the most efficient means of doing this is by the spiral-chute first introduced in 1896 in a breaker near Hazleton, Pa., by the writer. Another efficient device is the "waterfall-chute" first introduced in 1910 in the Lehigh Valley Coal Co.'s breakers, by Paul Sterling, Mechanical Engineer for that Company. It consists of a series of comparatively narrow floors in two vertical tiers, inclined toward each other and overlapping. Different dimensions are required for each size of coal. In its operation the coal flows from one floor upon the next floor below in the opposite tier, and so on, making the descent without material breakage. Either of these devices will deliver the coal to any vertical depth desired.

In like manner great savings can be effected with the exercise of due care in the design and construction of the loading devices. To illustrate the possible loss at this point in the process, it has been found by test that prepared sizes dropping 10 feet will depreciate 4% in fine chippings.

The preparation of bituminous and lignite coals must in a general way follow the same plan as that developed in the preparation of anthracite. This is manifestly so because the general formation of the beds and association of the impurities is the same. But because of the friability and soft nature of these coals the process has been modified to suit this inherent peculiarity. A close examination of the surface of the sized pieces of bituminous coal shows that the interlaminated bands of slate, although they may be as thin as india paper, protrude beyond the bands of coal, the coal having been worn or frayed away in the action of loading, dumping and screening. It is also observed that the very purest of pieces of bituminous coal have a greater angle of repose than even the least pure pieces of anthracite, showing that the slates are more silicious. It is plain therefore that the jig, because of the friability of the coal, would not succeed within economic limits. On the other hand because of the projection of the silicious interlaminated bands of slate beyond the surface of the coal the frictional machines have only a very limited range of efficiency. Any type of machine that produces impact will create serious disintegration and loss. Crushing the coal to half-inch mesh, and under, and treating it in a washer has proved very unsatisfactory and wasteful of the finest particles of coal, which have an equal value with the coarser particles in making coke, or for steam purposes and gas producing. Many ingenious schemes have been brought out to accomplish the coveted end. One scheme has recently been brought forward whereby the coal is treated entirely in the dry state, without the use of any water, and which recovers, it is claimed, about 98% of the values of the coal.

The market value of any coal depends primarily upon the number of heat units per pound. In the anthracite field the standards of purity are expressed in percentages of slate in the coal, this being a convenient and reasonably accurate way of expressing the purity of the coal. It is made possible to express it in this way because of the hardness of the coal and the sized condition of the prepared product. In the bituminous field, because of the presence of so much fine material in the product, the chemical analysis determining the heat units and percentage of ash, sulphur, etc., must form the basis from which the value of the coal is determined.

In designing a preparation plant the recognized scientific principles underlying all of the methods of preparation together with the individual peculiarities of the coal to be treated should unquestionably form the basis for formulating the scheme. If these principles are consistently adhered to a better preparation and more economic results will be obtained than can be obtained from trying to correct some inherent defects in the old methods, or to modify them beyond their fixed limits of efficiency.

INVESTIGATIONS RELATIVE TO THE BETTER UTILIZATION OF NON-COKING LOW-GRADE LIGNITIC COALS

By E. J. Babcock

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In the western portion of the United States there are extensive deposits of low-grade coals, largely in the form of sub-bituminous, non-coking and lignite coals. It is well, therefore, that our attention be called to the benefits to be derived from the proper utilization of our great western lignite deposits.

The extent and economic importance of these deposits is seldom recognized. In speaking of this kind of coal I may say that my own State, North Dakota, is one of the largest coal States of the Union, for it is estimated that this State alone has a coal area of over 32,000 square miles, capable of producing 400 or 500 billion tons.

When we stop to consider what these figures mean as to the immensity of these deposits in the West, it is not strange that our people ask why we are not deriving larger benefits from this great resource.

To the people of the West the economic utilization of our lignite, in whatever form it may be, is a matter of very great importance, and any new fields which promise to open up for the use of these lignite coals are well worth careful consideration.

For this reason there has been begun at the College of Mining Engineering of the University of North Dakota and Mining Sub-Station some extended work on the practical tests of a variety of methods of manufacturing briquets and producing gas from lignite, with a view to showing the utility and economy of these products for heating, for power and for lighting purposes. In addition to this considerable work has already been done with reference to other improved methods of burning and utilizing lignite coal.

The brief statements to which I must here confine myself will serve only as a summary of the work which has been done, the results thus far secured, and the promises which these results hold out for future development of our lignite fields. But what has been accomplished in our experimental work at the North Dakota School of Mines and Sub-Station leads me to believe that great improvements can be made in methods of burning and utilizing lignite coal and that the manufacture of gas and high-grade fuel briquets can soon be put on a successful commercial basis.

This will not only make our lignite coal much more serviceable and more generally used and of high enough value to be shipped for considerable distances, but it will also save an immense amount of slack coal which would otherwise be wasted, and which is estimated at from one-tenth to one-sixth of all the lignite mined.

In order to understand better why our lignite deposits have not been developed more rapidly, and to see what changes are needed in lignite to increase its value and usefulness, we may consider for a moment certain characteristics of lignite coal.

From the many hundreds of analyses of lignite coal made in our laboratories, the average composition of the dried coal would be about as follows:

Dried coal	Approximate average
Fixed carbon	52 per cent
Volatile matter	40 per cent
Ash	8 per cent

Moisture from coal fresh from the mine runs from 25 to 35 per cent.

In a general way, it may be said that the heating power of one ton of lignite of the quality mined in North Dakota when dry will equal about 65 per cent of a ton (1300 lbs.) of bituminous coal.

In appearance this coal is between a cannel coal and the brown lignite of Germany. It breaks up very easily when exposed to the air. This is no doubt due in large part to the rapid evaporation of water, there being usually from 25 to 35 per cent of moisture in the lignite as taken from the mines. The larger portion of this moisture escapes rapidly and causes, thereby, checking and splitting of the lumps into lignite slack. This is one of the greatest difficulties to be overcome in the commercial handling and utilizing of lignite.

In shipping coal containing so much moisture there is a heavy loss in freight and a corresponding reduction in the efficiency of the fuel. It is, therefore, important that the moisture be removed before shipment. But when this is done the fact that the coal still tends to become slack on burning presents another difficult problem. As a result, the utilization of lignite has been confined to comparatively narrow regions near the deposits. It would be an enormous stimulas to the utilization of lignite could it be satisfactorily and economically converted or *concentrated* into fuel free from moisture and of a size adapted to general commercial uses. These facts have directed our attention to the importance of some economical means of briquetting lignite.

At the present time, lignite coal is chiefly used in lumps in heating and cooking stoves and in engines for power. It is generally used in the most simple manner and very few special methods have been adopted for burning or utilizing this coal or for preparing it for the market. But there can be little doubt that for general stove and furnace use briquetted lignite would prove a most economic and profitable fuel. Lignite, however, has presented more serious difficulties to the process of briquetting than has bituminous coal. For this and other reasons the commercial briquetting of lignite has not, up to this time, been made a success in this country. I believe that a successful and sufficiently inexpensive method for the commercial briquetting of lignite is now being developed at the School of Mines and the Mining Sub-Station of North Dakota. The process which we are now working out is made commercially possible and profitable by operating the briquetting in conjunction with a gas-producing plant in which a variety of by-products are saved and utilized, and by which process the residue after the gas has been driven off can be successfully briquetted, thus concentrating and converting at once at the mine a low-grade lignite coal into a high-grade valuable fuel.

A large amount of work has been done by the speaker on the problem of briquetting lignite, its use in the production of gas and other means of its utilization. During the progress of this work, it has become clearly evident, by repeated experiments, that lignite coal contains two very widely different fuel constituents, namely, first, a large proportion of light gases, which pass off at a comparatively low temperature leaving, second, a residue, in character between coke and charcoal, which requires a very much higher heat for combustion and burns more slowly. This residue does not coke but remains as powder or slack which checks the draft and is slow in oxidation. The gases, however, are very light and are easily driven off from the coal at a comparatively low temperature. As a result, they are distilled off quickly, in the ordinary methods of burning, and are either burned with a flashy flame or frequently lost, in a high degree unconsumed, with the flue gases. Thus there is a very uneven combustion of lignite coal, and it is impossible with the ordinary type of furnace and fire box to secure a high degree of efficiency in its consumption. Following out the investigation suggested by these observations, the speaker has been led to inaugurate a systematic research on the production of gas from lignite coal and the utilization of the by-products. The first work along this line was of a preliminary nature and was carried on in a small way with laboratory apparatus in testing out a large number of both lignite and ordinary gas coals for the purpose of obtaining a general idea of the comparative quality and character of the gas, tar and residue, of the temperature required for most efficient work with the various coals and an estimate of the amount of ordinary by-products obtained from each coal under the condition employed.

One of the first problems to be studied was as to the uniformity of lignite relative to the gas contents, the character of the coke residue, the amount of moisture present, the tar, ammonia, and other by-products. To cover these points a large number of lignite coals were collected from different areas and these were subjected to a great variety of preliminary tests. The results proved conclusively that the gas from the lignites of different deposits is very similar, that it may be removed at nearly the same temperature and that it leaves a residue and by-products of a general similarity although the quantity of each varies considerably. The main products and by-products thus obtained were compared with those obtained from ordinary gas coals tested by the same processes. This preliminary work was so suggestive as to the possibility of the profitable production of lignite gas and briquets that the state made a special appropriation for establishing and operating a School of Mines Sub-Station in the heart of the lignite area.

These plants which have been designed and operated under my direction are of special construction, but are built upon well-known principles and, as far as possible, utilize standard machinery. Here both the briquetting and gas problems are being worked out on a scale so large as to ensure commercial results. The briquetting plant is capable of turning out ten tons of briquets per day, while the gas plant is a full size unit complete and of the standard type of construction, and consists of a bench of one retort, one condenser, one exhauster, two scrubbers, one purifier, one meter, all of the full size used in commercial plants, one gas holder, and miscellaneous apparatus necessary for the proper operation of the plant, such as meters, pressure gauges, recording thermometers, pyrometers, etc.

In the operation of the gas or carbonizing side of this plant, every step is carefully watched and made to approach as nearly as possible for a small plant the methods which might ordinarily be expected to be required for economic production. The experimental plant has given excellent results and indicates much wider possibilities as to the amount and quality of gas from lignite coal than was expected. There is generally used for each charge of the retort 300 to 400 pounds of air-dried coal, the moisture content of which is definitely known. The coal is crushed to an average size which would about pass through a two-inch ring. A series of experiments have been conducted to determine the temperature giving the largest quantity and the best quality of gas. It is found that the gas distills freely at 900 to 1000 degrees F., while the proper temperatures to give the maximum quantity and quality of gas seems to be between 1200 and 1500 degrees F. High temperature readings are made by the pyrometer along with simultaneous records of the rate of gas production. At frequent intervals gas samples are obtained for analyses. The desired heat is carefully maintained until the evolution of gas has ceased, after which the valves are closed, the non-coking residue drawn, cooled down, and carefully weighed. The gas is passed successively through the tar extractor, the condenser, the scrubber, the purifier, and finally to the gas holder, where the quantity is accurately read off from the holder after having been carefully corrected to normal temperature and barometric pressure.

After the gas is measured, analyzed and tested by the calorimeter to determine its heat value, it is used with different types of burners and in various practical ways to demonstrate its efficiency as a fuel gas. In the case of the by-products the tar is separated from the water and weighed and the ammonia separated and determined. The resulting proportion of non-coking retort residue remaining is also determined and an analysis made indicating the fixed carbon, volatile matter and ash. From this data the composition of the residue can be compared with that of the original lignite and also with that of bituminous and anthracite coal.

The following is a summary of yields of gas, residue and by-products and will represent a fair average from a larger number of determinations:

LIGNITE GAS ANALYSIS

(Considered average standard results of unpurified gas.)

Illuminants	Carbon Monoxide	Hydrogen	Ethane	Menthane	Nitrogen	Oxygen	Carbon Dioxide
3.11	17.24	45.16	0.14	15.59°	5.49	1.76	11.51

GAS YIELDS AND HEATING VALUES

Retort residue per ton of dry lignite, 1208 lbs.

Unpurified		Purified from Carbon Dioxide and Air	
Yield cu. ft. Calorific	Value in B. T. U.	Yield cu. ft.	Calorific Value B. T. U.
10,663	400.07	8,523	500.

Tar per ton of dry lignite, 20 to 30 lbs.

Ammonium sulphite per ton dry lignite, 14.5 to 15 lbs.

The gas made from the lignite has not only been subjected to a great number of chemical and calorific tests, but has also been put to a variety of practical tests. It has been burned in the ordinary manner for lighting purposes, and gives a most satisfactory and brilliant light with the ordinary mantle burner. It produces a clear smokeless flame and is easily regulated.

If the gas were purified to correspond with the treatment given the ordinary coal gas, the composition and heating value would be considerably modified. The heat units developed would probably be increased to between 450 and 500 B.T.U. as compared with standard gas of 600 B.T.U.

By this purification and complete removal of air a value of 500 B.T.U. could be obtained, but as already stated, the character of the gas and the freedom from objectionable impurities make unnecessary the additional expense of purification usually required of coal gas. The gas is dry and free from tar and other matters which would cause condensation or clogging. This is a material aid in the practical distribution and use of gas, especially in cold climates. The most severe and continued tests have been given to determine the condensation produced in carrying the gas in slightly protected or unprotected pipes during the cold weather. In this case no trouble was experienced from condensation.

The gas was used successfully not only for lighting but it gave perfect satisfaction and exceptionally high efficiency for power in gas engines. It is almost certain that this gas will produce a very cheap power for general purposes and for the production of electricity.

It is expected that much more extended tests along these lines for the utilization of this gas for power and the production of electricity will be carried on during the coming year. There is little doubt that the use of lignite gas for power purposes can be successfully employed and power produced at so low a figure as to exert a marked influence in the development of a variety of industries dependent upon cheap and abundant power thus available in the West.

The lignite tar is not adapted in itself, and without modification, to briquetting purposes. We have succeeded in using it very satisfactorily in small quantities by special methods of treatment which the investigations thus far carried on indicate as a possible method of utilization on a commercial basis. But this tar has a higher value as wood filler and for other purposes.

Lignite does not coke but remains in small shrunken fragments. This residue is high in fixed carbon and in excellent condition for briquetting.

We have carried on a large amount of experimental work in various methods of retorting and carbonizing lignite coal and have found that it carbonizes easily and gives very satisfactory results as to yield and quality in horizontal or vertical retorts, in special carbonizing ovens we have designed and in modified bee-hive ovens, in the lattter case, however, the yield is less and the gas is lost, thus reducing the profit.

The principles underlying the process of lignite briquetting which we have worked out at the University of North Dakota and the Mining Sub-Station depend upon the concentration of the raw lignite into a fuel of relatively high heating value and this concentration depends upon, first, the removal of the moisture; second, the expulsion and saving of the volatile gas; and third, the binding together of the concentrated residue into a strong, durable and satisfactory briquetted product.

After the coal has been sufficiently dried it is conveyed to the retort and the gas driven off as already explained. This process is usually completed in about four hours. At the end of this time the retort is discharged and the remaining coal residue quenched and crushed ready for the mixing with the binder. The coal and the binder are then warmed to the proper temperature and conveyed to the steam jacketed mixer where a very thorough blending of coal residue and binder takes place.

At this stage lack of uniformity and proper mixing produces a briquet which is uneven and of inferior quality. The temperature and the proportions of material must also be carefully watched. This is one of the most particular phases of the briquetting process. When the mixing has been carried on for a sufficient length of time the material is discharged and conveyed directly to the briquet machine.

Besides the conditions which have already been mentioned there are numerous details of mixing, tempering, etc., which can be properly understood and controlled only by experience. Conditions vary greatly with the kind and character of the binding material used.

The press used at the experimental plant is of the rotary type and is capable of producing 10–20 tons of 2¼ oz. briquets per day. It was built to order, especially for our Mining Sub-Station.

The briquets are discharged to a belt conveyor by which they are carried to the storage bins and the briquetting process thus completed.

Briquetted fuels have been extensively used in Germany and other parts of Europe where a variety of methods of manufacturing have been employed with varying degrees of success. With certain European coals briquet blocks have been produced without a binder, while in other cases lime, sugar wastes, starch, pitch and other binding materials have been employed. Numerous attempts have been made in this country to briquet coal without a binder by merely excessive pressure but this has proved unsatisfactory. By such a process the expense for power and operation nearly equals the additional cost of the binder. If raw lignite is briquetted under sufficient pressure to stand handling and weather action, it will still fail when fired under trying conditions, for when the briquet becomes red hot the gas expands and disintegrates the briquet to a greater or less degree. For these and other reasons no satisfactory briquets of American lignites have been produced on a commercial scale without a binder and even with a binder the raw lignite briquet is nowhere near as satisfactory a product to fire as the concentrated residue briquet.

Many hundreds of tests have been run by us with different proportions and under varying conditions, but up to the present time our best results have been secured in the use of a small amount of pitch as a binder base mixed with a small per cent of other materials, such as flour, etc.

Attention has already been called to the use of the gas in connection with the briquetting and to the revenue which may be derived for the plant by a combination of gas and briquetting plants, but in addition to this, as already has been pointed out, there is a considerable amount of ammonium sulphate formed (nearly 15 lbs. per ton of dry lignite) and tar, both of which have a ready market. Thus we see that by saving the various by-products and applying the revenue derived from them to the manufacture of the briquets the cost may be very materially reduced. In a large commercial plant the saving of a few cents per ton will be sufficient to net a good profit.

The briquets which we are now able to run out regularly and in considerable quantities are first class in every respect. They are strong, hard, durable and will stand weathering excellently and burn with entire satisfaction in most any type of stove or furnace. They have been tested in a very great variety of ways and have always made the best showing.

The chief advantages to be gained in the briquetting of lignite are the large gain in heat value, the prevention of slack so characteristic of this coal, the holding together of the mass during combustion, the prevention of matting in the firebox, free burning, uniformity in size, cleanliness in handling, and decrease in the loss, caused by the dusting. In addition to these points briquets can be used to better advantage in a variety of stoves, and may be kept under more perfect control than the ordinary form of lignite. But the greatest advantages are the increased heating power and the fact that the lignite is put into a satisfactory form which will stand weathering, handling and burning without disintegration.

There is comparatively little loss through the grate bars, the briquets burn to a perfect ash in which practically all of the fuel has been consumed.

The calorific determinations which are constantly made show about the following averages:

Lignite as mined	7,500 B.T.U.
Lignite briquets	11,500 to 12,000
Anthracite coal	12,000 to 13,000

It will be seen that lignite briquets have raised the heating power of the lignite about 70 per cent and give a calorimeter value of approximately twelve-thirteenths the heating value of anthracite coal. The chemical analysis of the briquets as compared with a raw coal is shown by the following table:

CHEMICAL ANALYSIS

	Moisture	Vol. Matter	Fixed Carbon	Ash
Lignite as mined	35.01%	25.11%	34.67%	5.21%
Lignite briquets	0 to 6%	2 to 8%	75 to 85%	10 to 14%

A great variety of heating and evaporative tests of a very severe and practical character have been made which indicate that the lignite briquets thus prepared are fully equal in heating and evaporative tests to the higher grade coals.

One and one-half tons of lignite will produce about a ton of these briquets in addition to 12,000 or more cubic feet of gas. The briquets have about twelve-thirteenths the actual heating value of average anthracite and can be made at a sufficiently low cost to put the industry on a commercially satisfactory basis in at least some of our Western States.

Considering the ease with which the gas is produced, the low price of the original lignite and the value of the residue, this gas should have a large commercial value for heating, lighting, and power purposes, because of the low price for which it could be sold if manufactured in a plant used to produce briquets from the residue.

No detailed statements of the cost of operating a large commercial plant are given in this paper, for the reason that the cost per ton of briquets and per thousand feet of gas and other by-products will depend upon a large number of factors, any one of which may materially affect the cost.

In addition to this, the plant could be operated under many modifications of the general process which has been explained; for example, all or a part of the gas might be sold for heating or lighting purposes or converted into electricity. The extraction and production of gas might be carried further in one plant than in another, or the by-products such as tar, ammonia, etc., could be recovered and marketed, used in part, or entirely neglected. All of these conditions would materially modify the cost of production.

From the results obtained by the methods being worked out at the School of Mines and Sub-Station of the University of North Dakota, there seems little doubt but that the briquetting and the production of gas from lignite will in the near future be put on a commercially satisfactory basis, at least in the State of North Dakota. This will prove of great value to those communities nearest the great lignite deposits in the West. In some of these the wastes can be converted into electricity which in turn could be sent to surrounding towns and villages, thus distributing power and light from numerous central power plants. Such an arrangement will not only be a great saving of our fuel resources but will also result in establishing many industries which can be developed by abundant and cheap electric power.

In addition to this will be the production of briquets of high fuel value and so uniform and desirable that the product can find a ready market. The best utilization of our vast deposits of low-grade lignite coals is a problem of very great importance to many of our Western States, one which will mean, in the aggregate, the saving of large sums of money and aid the introduction and development of a variety of manufacturing industries, while at the same time it will illustrate the value of scientific and technical work in the economic and industrial development of a community, a state or a nation.

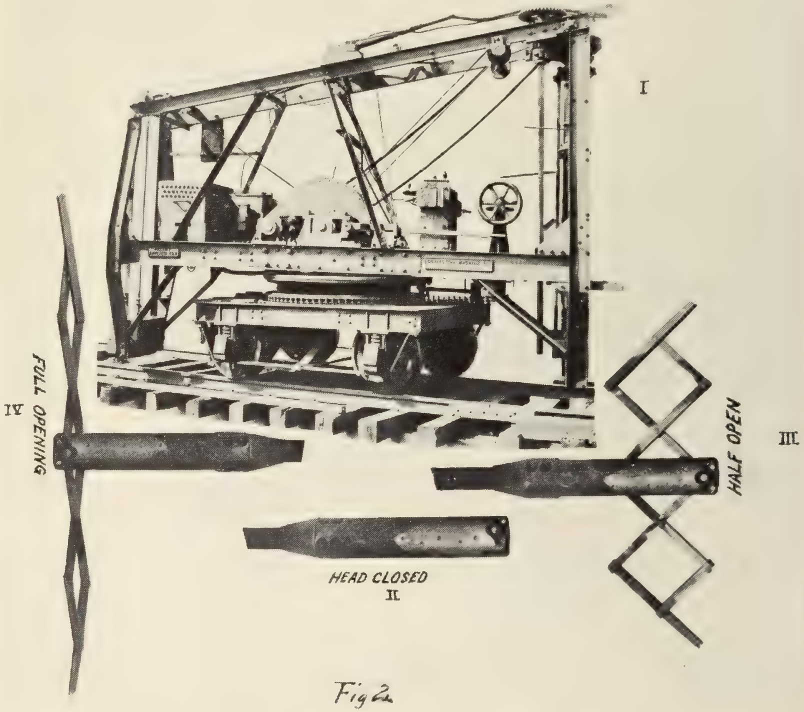

Fig 2a

THE BEEHIVE COKE OVEN INDUSTRY OF THE UNITED STATES

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The manufacture of coke in the United States according to authenticated reports was begun about 1817. From this date on, mention is made from time to time of the use of coke for metallurgical purposes, but the coke was made on the ground in pits or mounds and no record of any coke made in ovens can be found until the year of 1841 when two carpenters and a stone mason formed a partnership for the building of two ovens and the manufacture and sale of coke. This plant was built in the famous Connellsville region, and although the business venture was unsuccessful the coke proved useful for foundry purposes. This venture, together with experiments carried on during the next ten or fifteen years, fully demonstrated the value of this fuel, and production increased by leaps and bounds into the vast beehive coke oven industry which completely dominated the field until 1893, when the first coke oven plant for the recovery of by-products was introduced into this country.

The evolution of the modern beehive oven started as shown above and the process of coking in this type of oven has not materially changed, the modern increased efficiency being due in great measure to improvements in the ovens and the preparation of the coal before charging into the ovens.

BEHIVE OVENS

The beehive oven in its essential details may be described as a circular vaulted fire-brick chamber constructed on a suitable foundation, with flat tile bottom, an opening in the top through which the coal is charged and the products of combustion escape, and an arched door at the bottom about three feet high, through which the air for combustion is admitted and the coke watered and drawn. The many variations and improvements since the ten-foot-diameter oven of the carpenters and stone mason of 1841 have led to the present standard beehive oven as shown in section figure 1. The size of the door and the trunnel-head, height of dome and diameter of oven vary in different localities, but the essential features are the same. The ovens are built in single rows called bank ovens or in double rows back to back or staggered. The coal, the amount of which is previously determined as the oven is to be burned 48, 72 or 96 hours, is charged into the oven from a larry car operated on a track above the ovens. The coal in falling into the oven forms a conical pile and is leveled by means of a scraper operated by hand. The door is bricked up to within two or three inches of the top and the oven left until the heat held by the bricks from preliminary heating, or the previous charge, raises the temperature to the point where the volatile matter, distilled from the coal, finally catches fire. The coking proceeds from the top downwards and the burning of the volatile to maintain the required heat is regulated by closing up the air opening as the amount of the volatile falls off. After the volatile has ceased to come off the process is finished. The door is then torn down, the coke watered on the inside of the oven by means of a spray of water and is then drawn from the oven.

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The beehive oven arranged for the mechanical drawing of coke differs from the above only in the width of the door and projecting iron jams at the sides of the door. Mechanical drawing of the beehive oven has been resorted to on account of the scarcity of labor and not from any increased efficiency resulting from the mechanical operation *per se*. There is reason to doubt if mechanical drawing shows any material reduction in the cost of operation when all the items are taken into consideration. It undoubtedly breaks up the coke more, producing a large percentage of breeze, but, on the other hand, it reduces the time of drawing ry materially. The enlarged size of the door makes drafting more difficult and leads to much burned coke. The practice of watering far in advance of drawing and especially where the precaution of putting the lid on trunnel-head is not followed, leads to the rapid cooling of the oven and reduces the yield by increasing the length of time necessary for subsequent charges to ignite. This increases the time to burn the same size of charge or necessitates the reduction of the charge to burn ovens down on time.

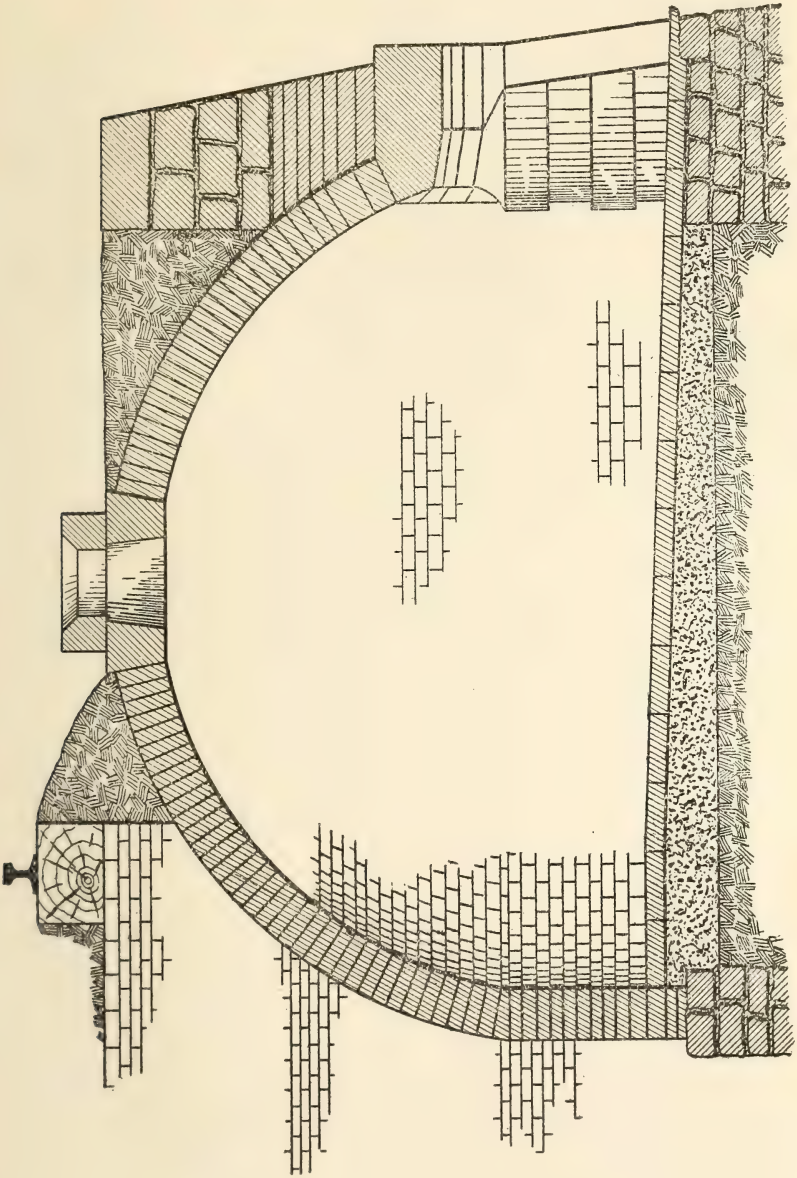


Figure 1—Beehive Oven Section of Standard Type. 2′ Diameter 7 6″ High

Lately a patented machine for the mechanical leveling of coal in beehive ovens has been put on the market and is now in operation in the Connellsville coke region (Figure 2). It runs on the larry track, uses the larry trolley, and operates through the trunnel-head. This machine gives evidence of proving its usefulness, not only by the elimination of hand labor, but by encouraging the proper leveling of the ovens, an operation too often neglected or considered of little importance. The proper leveling of the coal in any oven is a matter of great importance, and it is a source of regret that so little attention is paid to this feature of the process by the beehive operators throughout the country. With improper leveling, the different heights of charge become coked to the bottom at different periods of time, thus exposing the top surface of the coke to the prolonged action of the air admitted for the completion of the process, with a resultant loss of coke, or if the process be stopped short of completion the coke from the higher portions exhibits black butts.

LONGITUDINAL OVENS

The decreasing efficiency and scarcity of labor and the increased cost of coking coal has led to experiments to reduce both of these factors, first by making the coking operation non-dependent on large numbers of laborers, and secondly by decreasing the cost of actual operation. From these experiments was evolved in 1906 the longitudinal oven (a modification of the old Belgian type), with its mechanical devices for leveling, pushing and loading of coke. Figure 3 shows a section of this type of oven and the method of operation. In its essential details it may be described as a long, narrow, rectangular chamber generally somewhat larger at the discharging end, with sloping barrel roof approaching the center from both ends, a trunnel-head in center of roof, flat tile bottom and doors the whole width of the chamber at either end. These ovens are placed side by side forming a block and are charged as in beehive practice, from a larry running on a track supported on the ovens. The oven is drafted from both ends, after the method of the beehive oven, and the coke, after being watered on the inside of the oven, is pushed out by means of a pusher, devised for the purpose, onto a traveling conveyor, which transfers it to cars;

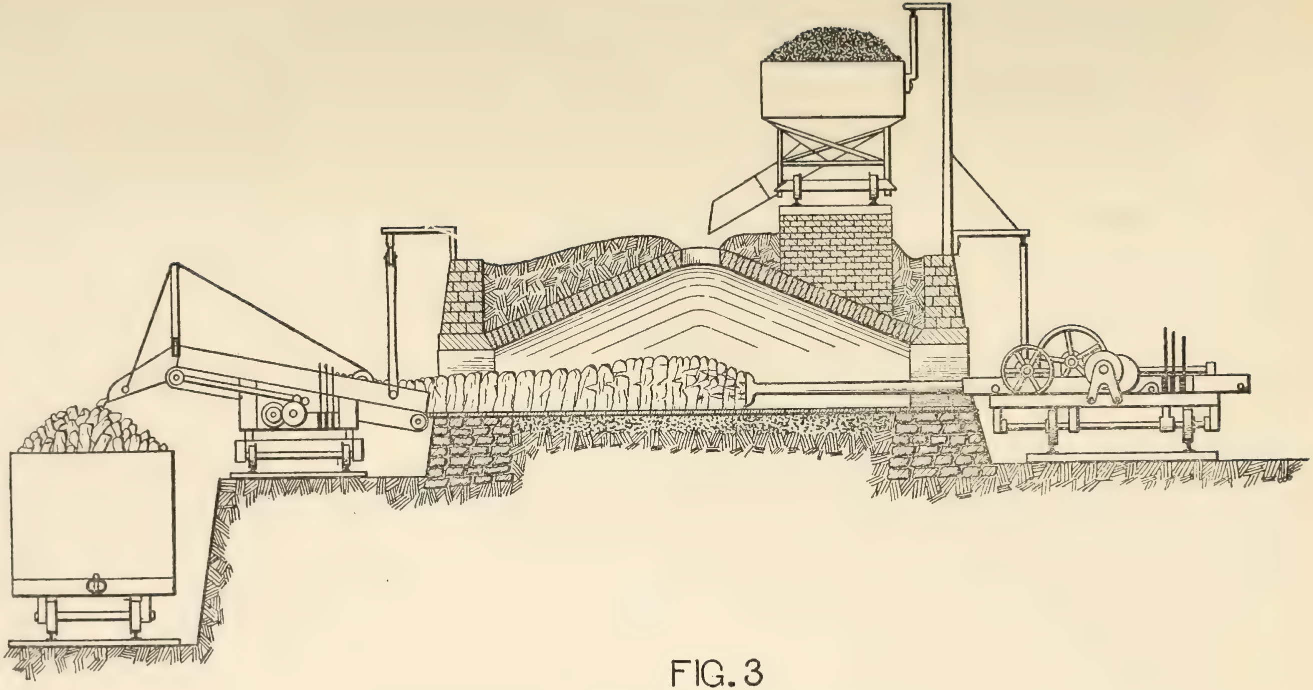


FIG. 3

LONGITUDINAL OVEN

SECTION SHOWING METHOD OF OPERATION

screening being more or less fully accomplished during the passage of the coke along the conveyor. During the last three years this type of oven has come into prominence, especially in the Lower Connellsville region, and many claims are made for it in regard to increased yield, better product, lower cost of production, etc., but judgment must be withheld until these ovens have been in use for a longer period and prove their worth by actual service.

BEEHIVE OVEN WITH ARRANGEMENT FOR UTILIZING WASTE HEAT OF GASES

Waste heat from beehive ovens is utilized at some few plants in this country for the generation of steam. The ovens, either bank or double-row, are provided with a tunnel, built in case of the bank ovens directly behind, and the double-row between the rows, and extending the whole length of the ovens to be used for this purpose. From each oven a flue is provided leading from the oven to the tunnel and so arranged that it can be dampered off. This flue is placed in the crown of the oven above the height to which the coal is charged. Numerous schemes for placing the flues leading from the oven to the main flue have been tried. The usual practice is a short straight one from the oven direct to the tunnel. At the boiler end a large stack is placed to cause proper draft. In operation the regular trunnel-head is covered and kept closed, the gases passing through the flue into the tunnel instead of into the open air as in the case of the ordinary beehive practice. The waste heat passes along the tunnel in the direction of the draft chimney and through the boiler setting before passing out of the stack. The temperature in these flues is very high (around 2700° F.) and the building of flues to withstand this heat is a matter of much consequence. The temperature of the gases at the boiler approximates 2000° F., and the gases are sufficient in amount to generate from 12 to 20 horse power per oven. The ovens burn hotter under these conditions due to the better draft on the ovens than when burning into the air.

BEEHIVE COKING IN THE SEVERAL COAL FIELDS OF THE UNITED STATES

Beehive coke is produced in five of the seven great coal fields of the United States: Appalachian field in Pennsylvania, Virginia, West Virginia, Ohio, Tennessee, Georgia, Alabama and Eastern

Kentucky; the Eastern Interior field in Illinois, Indiana and Western Kentucky; the Western Interior field in Kansas, Missouri and Oklahoma; the Rocky-Mountain field in Colorado, Montana, Utah and New Mexico; and the Pacific Coast field in Washington.

Pennsylvania has from the beginning of coke making in the United States maintained the supremacy as to production, and the beehive oven coke from the Connellsville region is still considered the standard coke of the country. All other cokes from whatever section of the country are judged by comparison with this standard. The coal from this region, with approximately thirty-one per cent of volatile matter, seems to contain just the proper amount and composition of this volatile and to be given off at the proper temperature and time to produce a maximum yield of coke with a minimum loss of fixed carbon during the process. Practically all the coal in this region is mined for the production of coke and is charged into the ovens without any special preparation whatever. About fourteen per cent of the coal from other coking regions of the State is washed before coking, and the coke produced is of varying purity and physical structure. The table herewith shows the range of composition of the Pennsylvania cokes:

Moisture	.23 to .91
Volatile matter	.29 to 2.26
Fixed carbon	92.53 to 80.84
Ash	6.95 to 15.99
Sulphur	.81 to 1.87

West Virginia coal is of exceptional purity and the coking industry, although second in production, is mainly as an incident to the furnishing of steam and domestic coal. More than sixty per cent of the coal charged into the beehive ovens is slack and less than ten per cent of it is washed. At those plants where proper attention is given to preparation of the coal and subsequent operation of the ovens themselves, a coke of superior chemical and physical properties is produced. The quality of the greater amount of West Virginia cokes is exceptionally good, but a wide variation is still to be found in the chemical composition of the cokes of this State as the following range shows:

Moisture	.07 to .60
Volatile matter	.46 to 2.35
Fixed carbon	95.47 to 84.09
Ash	4.00 to 12.96
Sulphur	.53 to 2.26

The coke produced in Virginia is from coal in the southwestern part of the State and is all from unwashed coal. In chemical and physical properties it resembles the cokes from southern West Virginia.

The cokes from the southern part of the Appalachian field, comprising the States of Tennessee, Georgia, Alabama and Eastern Kentucky, are for the most part made from washed coal, the quality of coke is poorer and the ash and sulphur are high. The average ash is in the neighborhood of 14 or 15 per cent, and often running up to 16 and 18 per cent where proper attention is not paid to preparation and coking. The Birmingham district of Alabama produces a fairly large amount of good beehive coke, with an ash content averaging 11 per cent, but such coke is the exception and not the rule.

Range of composition of above coke:

Moisture	.75 to 1.34
Volatile matter	.75 to 1.95
Fixed carbon	91.20 to 77.81
Ash	7.30 to 18.90
Sulphur	.58 to 1.77

Beehive coking operations in the States of Illinois and Indiana are practically at a standstill. A few ovens for experimental purposes have been built in these States and have demonstrated the practicability of producing coke from some of these coals. The coking possibilities of Illinois and Indiana coals is confined to small areas, and beehive operations will hardly be established in these fields.

Kansas, Missouri and Oklahoma have had poor success in the production of coke and the total output from these States is very small.

Practically all the coke from the States of Colorado and New Mexico is made from coal previously washed and crushed before charging into the ovens. A plant of 350 beehive ovens of concrete construction is one of the novelties of this region. The ovens proper are of the ordinary beehive type, 13 feet in diameter and 7 ½ feet in height, constructed of firebrick and tile. The ring walls and wharf walls are of plain concrete, the battery walls and larry track columns of reinforced concrete. The yield of coke is reported above the average for the region and is believed to be due in great measure to the concrete construction which prevents the entrance of excess air so common in cracks developed in ordinary beehive construction. For details of this plant the reader is referred to Mines and Minerals, February, 1910, pages 429 to 432. At another plant in this region the ovens are provided with underflues which convey the gases from the coal under the oven bottom to a large flue, back of the ovens, which carries it to the powerhouse. This power plant furnishes all the power for operation of the mines, for ventilation, electric haulage, coal washing and crushing, and steam heat for the company buildings and electric lighting for the entire community. The flues under the oven bottoms serve to hasten the coking and prevent black ends. The operation requires careful watching to prevent the coal from coking up from the bottom as well as down from the top. When the bottoms become too hot and this coking upwards does take place a distinct line of demarcation, where the two operations meet, is seen over the whole charge of coke.

The cokes of these States are uniformly low in sulphur, scarcely ever exceeding .75 per cent, but the ash is high, averaging 16 per cent.

The coals used for manufacture of coke in Utah are practically identical in character with those of western Colorado, but exhibit the surprising characteristic of losing their coking properties and becoming non-coking if kept in storage for any length of time.

Montana produces a very small amount of coke, all of which is used in copper smelters. Attention is called to the fact that the only restriction placed on the coke is that the ash content shall not exceed 20 per cent. This restriction is only complied with when the coal is washed. An average analysis of this coke shows 18.00 to 21.00 per cent ash and over 2.00 per cent sulphur.

Washington is the only State west of the Rocky Mountains that contains coking coal and the area within the State is small. All the coke is made from washed and crushed coal and is of fairly good quality. An average analysis of this coke follows:

Moisture	.92
Volatile matter	1.50
Fixed carbon	59.58
Ash	18.00
Sulphur	.52

THE BURKHEISER WET SULPHURETTED HYDROGEN, CYANOGEN, AND AMMONIA PURIFYING AND RECOVERY PROCESS

By Karl Burkheiser,
Engineer, Hamburg

The object of the Burkheiser process is to absolutely purify coal gas and to combine the sulphur contained therein with the ammonia, so as to form ammonia salt. In these fundamental efforts the Burkheiser process differs from all other processes which try to attain the combination of the sulphur in the gas with the ammonia.

The Burkheiser process is not only a process for the recovery of by-products, but also a good gas purifying process, which is superior to the processes used up to now for gas purifying.

In these efforts, the author started from the principles used up to now for the sulphuretted hydrogen purifying by the use of bog ore. But he submits the bog ore before its use to a preparation by roasting, and thereby removing all the organic parts, and also by withdrawing from it part of the chemically combined waters. From the original bog ore ($Fe_2(OH)_6$) is formed a red modification, poorer in water (probably from the combination $Fe_2O_3H_2O$), which has a great absorbing capacity for the sulphuretted hydrogen. By so doing, it is not only possible to confine the gas purifying itself to the smallest imaginable space, but also to give the purifying mass a form which gives, in the oxydation of the iron sulphide combinations to SO_2 and SO_3, an absolutely pure SO_2 and SO_3.

The fundamental principles of the process consist in short of completely removing, by means of the above-mentioned purifying mass, the sulphuretted hydrogen from the gases, and in the regeneration of this purifying mass to drive the oxydation up to sulphur di-oxide and sulphur tri-oxide, and then to wash out by one and the same wash fluid the so-formed sulphurous acids, as well as the ammonia of the gases, without the carriers of these two products coming into contact.

At the beginning, the author carried out the process in such a way that by means of his purifying mass, which (in order to diminish the pressure) he used in pieces of 10 to 15 mm. diameter, and placed in purifying tanks of 1½ to 2 meters high, where the purification from sulphuretted hydrogen took place, and by regenerating the thus formed iron sulphide combinations in the purifying tank itself by introducing a regulated air current up to the formation of sulphur di-oxide and sulphur tri-oxide. But very soon difficulties were met with in the procuring of this product in pieces of a certain size, and also with the increase of the size of the plants the difficulties increased in carrying out the regenerating of large quantities in the purifier itself. In the construction of the purifiers, not only had to be considered their function as a sulphuretted hydrogen purifier, but also their function as a regenerator, whereby complicated constructions, with all their disadvantages, resulted; besides, the careful regulating of the temperatures made the supervision of the plant difficult.

To remove these difficulties, the author introduced the wet sulphuretted hydrogen purifying which has for such a long time been considered the ideal in the gas industry.

For nearly 50 years they have occupied themselves with this ideal of gas purifying, and the carrying out of the sulphuretted hydrogen purification in a wet way. Of the many proposed methods, only the exceedingly interesting process of the Englishman Claus succeeded (German Patent No. 23763, Class 26) and which was used for some time in practice. Claus intended to remove the sulphuretted hydrogen of the gases by washing with ammonia, and to regenerate the thus formed solution of ammonium sulphide and ammonium carbonate. The exceedingly high contents of carbonic acid of the gases, however, necessitated the ammonia liquor continuously in motion being about six times the amount the gas contained originally, so that the ammonia loss was such as to prevent successful working.

Latterly they tried to wash out the sulphuretted hydrogen with the help of sulphurous acids by impregnating tar oils with sulphurous acids, and treating the gas with this wash mass according to the form $2\ H_2S + SO_2 = 2\ H_2O + 3\ S$. But it was soon found that the reaction only took place to a small extent by these means, and that the washing of the gases was a very imperfect one.

After the efforts of the author had become known, other inventors took up the idea of using the sulphuretted hydrogen of the gases for the formation of ammonia salt, and an attempt was made to wash out the sulphuretted hydrogen by Metallic thiosulphate, and to change the thiosulphate into poly-thiosulphate, and finally into sulphate. Experiments were made with a large number of metals such as zinc manganese, iron, etc., and latterly also with ammonium poly-thionate. In spite of the exceedingly numerous methods which were used, success has not yet been attained, and probably for the reason that the full reaction with thiosulphate and poly-thionate only takes place in rare cases, and principally in a certain direction, and also because these reactions are always affected by a large number of secondary reactions. But even if such reactions are successful quantitatively, there is always *only so much sulphuretted hydrogen removed* from the gas as corresponds with the contents of the ammonia in the gas, so that a large portion of the sulphuretted hydrogen remains in the gas.

Such processes are always only sulphur combining processes, and not sulphur purifying processes, because, as mentioned above, apparatus for the removal of the surplus sulphuretted hydrogen is necessary in just the same manner as in the process up to now.

How the author carries out in the wet process the complete purifying of the gas from sulphuretted hydrogen, cyanogen and ammonia, and at the same time binds the ammonia salt, and uses the cyanogen products, is shown in the process described hereafter, and in the drawing attached.

The gas coming from the ovens is freed from tar and cooled only so far as is necessary for a rational tar separation; the condensate falling out in this part of the plant is distilled in the usual manner. The vapours containing NH_3 from the distilling column are again conducted to the gas current so that the whole of the ammonia, the whole of the sulphuretted hydrogen, and all the cyanogen of the gases is contained in the gas entering the purifier.

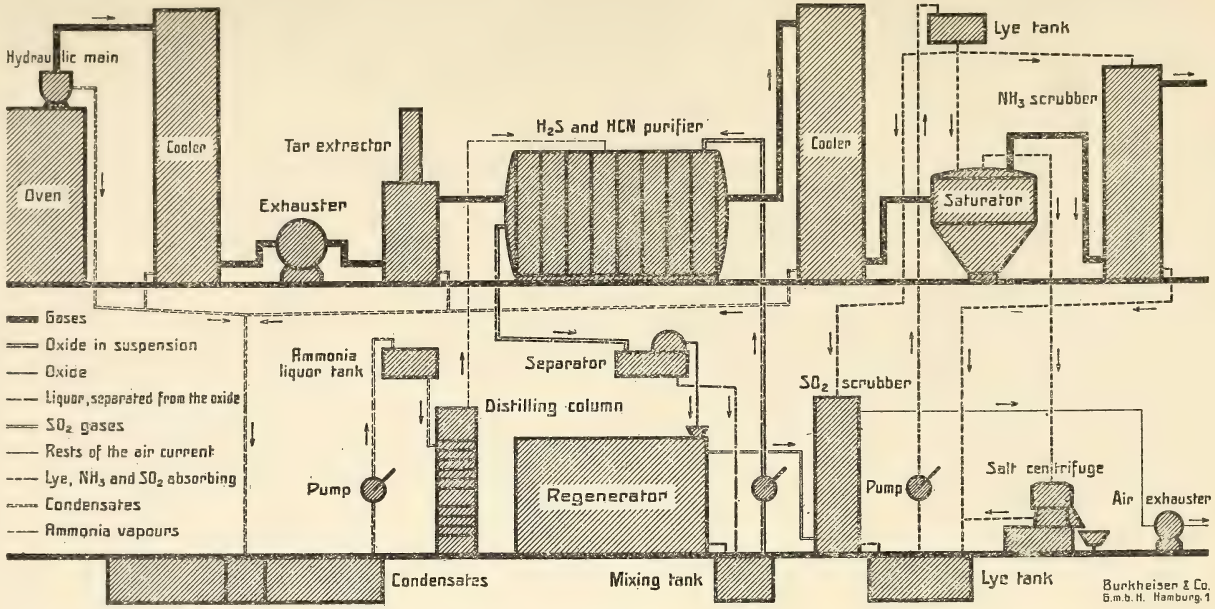
The gas now freed from tar, but containing all its ammonia, sulphuretted hydrogen and cyanogen, goes into a rotating sulphuretted hydrogen and cyanogen purifier which is an ordinary "Standard" washer, and is divided into a number of chambers connected together.

Into this rotating purifier the wash fluid is sent, which consists of very fine "Burkheiser" purifying oxide mixed with water. This wash fluid is kept in motion as it passes through the purifier, by means of purposely constructed boards and shovels, so that an intimate contact of the gas with the wash fluid, takes place. The supplylng of the purifier with the wash fluid (oxide in suspension) takes place from a mixing tank with revolving stirrers. The wash liquid flows from chamber to chamber of the purifiers, through which the gas goes in a counter current, so that in the first chamber in the direction of the gas current, in which the impure gas enters the purifier, the product is nearly saturated with sulphuretted hydrogen, the ammonia contents of the gases serving as a reaction carrier. At the beginning of the process, the wash fluid absorbs ammonia from the gases. A loss of ammonia however, does not take place, for, as will be shown below, always the same wash fluid saturated with ammonia is in circulation.

The oxide in suspension absorbs the H_2S by forming iron sulphide compounds, and at the same time absorbs also all the cyanogen of the gases. There are formed in the presence of sulphur, Alkali polysulphides in the ammoniacal liquor, which carries along the oxide, and these absorb the cyanogen of the gases. There are thus formed alkali-sulpho-cyanide compounds, which, as they are very easily dissolved, are easily separated from the iron sulphide combinations.

The cyanogen purifying takes place therefore without any special arrangement, in such perfection as is not obtained in any cyanogen purifying process used up to now, which, on account of its complicated apparatus, etc., has made economical working very difficult. The cyanogen purifying therefore not only costs nothing, but is also more perfect, and besides, the cyanogen is obtained in a form which, as explained below, is changed to a valuable by-product, that is, ammonia. There thus disappear in this process, not only the expensive apparatus for the cyanogen

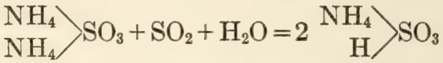
BURKHEISER'S „WET" PROCESS.



purifying, but also, at the same time, the cyanogen itself with all its drawbacks, and in its place is given an increase of the ammonia yield.

The gas coming out from the purifier contains now only ammonia which is absorbed after the principle of the author, that is, with a second lye continuously circulating through the saturator and ammonia washer. The wash fluid which is brought into contact alternately with the gas containing NH_3 and the air containing SO_2 consists of a mixture of neutral and primary sulphite, and is therefore capable of absorbing NH_3 as well as SO_2 gas.

$$\left.\begin{matrix}NH_4\\H\end{matrix}\right\rangle SO_3+NH_4=\left.\begin{matrix}NH_4\\NH_4\end{matrix}\right\rangle SO_3$$



$$\left.\begin{matrix}NH_4\\NH_4\end{matrix}\right\rangle SO_3+SO_2+H_2O=2\left.\begin{matrix}NH_4\\H\end{matrix}\right\rangle SO_3$$

In other words; if one sprinkles the ammonia washer and saturator with a wash fluid of primary and neutral ammonium sulphite, the neutral sulphite of the wash fluid will pass this apparatus unchanged, whilst the primary ammonium sulphite changes into neutral ammonium sulphite. On the contrary, the ammonium sulphite will remain unchanged by the sprinkling of the SO_2 washer, and the normal ammonium sulphite will change into primary ammonium sulphite. The wash fluid collected in the low tank which comes from the NH_3 and SO_2 washer, consists therefore again of a mixture of primary and normal ammonium sulphite. That this mixture of primary and normal ammonium sulphite is produced in the working by itself, is specially clear if one follows up the process from the beginning and uses water as a wash fluid. If one begins the wash process with pure water and sends it through the ammonia washing apparatus, the wash fluid running off from this consists of ammonia water (NH_4OH). If one sends this ammonia water to the SO_2 washer, the wash fluid running off from this consists of primary ammonium sulphite ($NH_4OH+SO_2=\left.\begin{matrix}NH_4\\H\end{matrix}\right\rangle SO_3$). In the further circulation this lye goes again to the ammonia washer where it absorbs NH_3, so that normal ammonium sulphite is formed $(\left.\begin{matrix}NH_4\\H\end{matrix}\right\rangle SO_3+NH_3$

$= \left.\begin{matrix} NH_4 \\ NH_4 \end{matrix}\right\rangle SO_3$). If one pursues this circuit further, one finds that continuously takes place the formation of primary and normal ammonium sulphite. If one now runs off the wash fluid from all the apparatus into a deep tank, and pumps this to a high tank, the result is the above-mentioned mixture of primary and normal ammonium sulphite. This characteristic composition of the wash fluid, which of course can vary to one or the other side, is easily obtainable with all gases. All coals used for gas making have considerably more sulphur than is necessary for the combining of the ammonia. One is therefore quite at liberty to give more or less sulphurous acid to the wash fluid. From one molecule $\left.\begin{matrix} NH_3 \\ NH_4 \end{matrix}\right\rangle SO_3$ there is formed two molecules $\left.\begin{matrix} NH_4 \\ H \end{matrix}\right\rangle SO_3$, whereby the lye is always more concentrated until the saturating limit is obtained, when the normal ammonium sulphite falls out of solution as its solubility is less than primary ammonium sulphite. This takes place in the saturator where the gas with its whole ammonia contents passes through the wash fluid, and gives up the greatest part of its ammonia, whilst in the ammonia scrubber the remaining small part of the ammonia contents of the gases gives a small enrichment to the wash liquid. By this is obtained a completely perfect washing out of the ammonia, as only a part of the efficiency of the wash fluid conducted to the ammonia scrubber is required.

The total sulphuretted hydrogen and ammonia purifying consists therefore only of a rotating purifier, a scrubber, and an ammonia final washer. Of special importance in the process is that the dimensioning of the sulphuretted hydrogen and cyanogen purifier is independent of the sulphur contents of the coal in use, because the wash liquid (mass suspension) flows continuously through the purifier. It only requires a more or less quick pumping of the wash fluid through the purifier in order to render the purifying a complete one. The purifier itself is divided into six-eight chambers, and is so arranged that already in the fourth chamber (reckoned from the gas entrance) the gas is completely purified of sulphuretted hydrogen and cyanogen.

As is seen, the whole gas purifying takes place continuously in this wet process without any interruption, and without any change of valves. It does not require any stopping or diversion of the gas current in order to take out the saturated product from the purifier. The mass in suspension flows out automatically through overflows from chamber to chamber of the purifier, and the gas current goes without interruption always the same way. The saturated product is, so long as it remains in apparatus containing gas, always in suspension, whereby all dangers which were connected with the usual sulphuretted hydrogen purifying process are removed.

As is seen from the above, the regenerating from the used product, and the production of the sulphurous acid, does not touch any phase of the actual purifying process. The outflowing mass in suspension from the purifier which contains the purifying mass saturated with sulphur, and in its liquid, sulpho cyanide compounds is conducted by a natural fall through pipes into a special space, completely separated from the gas purifying plant, and here the mass is separated from the liquid. The mass separated from the liquid falls into a regenerator; the liquid flowing back into the mixing tank. The regenerator consists of two drums placed horizontally one above the other, in each of which there are rotating arms which move the mass continuously, and at the same time carry it through the drums. The mass separated from the fluid, and still a little damp, falls through a shoot first into the top drum of the regenerator, and is revivified here only up to the production of free sulphur, and is dried on its passage through the drum by the reaction heat. The dry product falls again automatically into the drum lying underneath in which the reaction is proceeded with up to sulphurous acid. These SO_2 gases go into the air washer. The product coming from the regenerator is completely regenerated and falls into the mixing tank, and begins its circuit again.

As observed above, the liquid separated from the saturated product contains the Alkali sulpho cyanide compounds which are changed into ammonia. This takes place in such a manner that from time to time a part of the wash fluid overflows into a special tank and is mixed with lime, and calcium cyanide formed, the free ammonia being led back again to the gas. This calcium cyanide is exposed to a high temperature, whereby the sulpho cyanide is completely changed into ammonia, which also is conducted to the gases. Very minute tests of the author have shown that a complete conversion of the sulpho-cyanide compounds to ammonia is obtained by a reducing and oxydising substance and that this fact has an important bearing on the varying productions of ammonia and cyanogen obtained in the distillation of coal. Therefore not only a cyanide purifying at low cost is obtained, but with the most simple apparatus, and the most simple means, all cyanide is changed into ammonia, whereby, according to the cyanide contents of the gas, the ammonia yield is increased by 10% and more.

The amount of product (oxide) which is necessary for the sulphuretted hydrogen absorption in this process has no comparison to the dry purifying process. For the purifying of a daily gas production of 350,000 cubic feet, about 1,000 lbs. of product suffices. In one hour therefore, only about 40 lbs. of product pass through the regenerator, whilst in the ordinary purifying for the same gas quantity, 40 tons product would have to be put into the purifiers. The amount of product present in the regenerator is therefore exceedingly small, so that the regenerating process can be carried out under the most favorable conditions. The product is always again completely regenerated. A renewal of the purifying product is not required except to replace the natural losses in the working.

A further advantage compared with the dry process is the use of the product in the form of dust; the drawbacks due to the breakage of a used lump product, through the influence of chemical reaction by repeated regeneration, is completely done away with, and the life of the product is not influenced by its continual use.

The salt outfall from the process consists of a mixture of ammonia sulphite and sulphate, firstly by production of SO_2 as well as SO_3 in the regeneration, and secondly because oxydation takes place during the circulation of the wash fluid. In the space of a few weeks, however, all sulphite which is contained in the salt, oxydises to sulphate without any loss of ammonia. The salt has always a high percentage of ammonia, is of a beautiful white colour, and contains no free acid, and the corrosion of sacks so often found with sulphate produced with sulphuric acid, is absent.

The advantages of the process are chiefly as follows:

1. The saving in capital cost of plant owing to a very great reduction in the requirements for space of the purifying plant, and the small cost of the apparatus and buildings.

2. The increase of working profits by the greatest use of raw materials; and that:

a. By uniting the purifying and ammonia recovery to a single and simple self-contained process.

b. The cyanogen purifying is carried out in a perfection not obtained before, without any extra cost or special arrangements, at the same time as the sulphuretted hydrogen purifying.

c. For the ammonia recovery, the introduction of foreign sulphuric acid is completely replaced by the sulphuretted hydrogen contained in the gases.

d. The cyanogen nitrogen is changed to a valuable ammonia nitrogen, and results in an increase of 10 to 15% of the ammonia output.

e. An increase of the life of the apparatus is obtained. The apparatus does not require any lead work, as no acid is used, but in its place a wash fluid which does not cause any corrosion of the apparatus.

f. The large requirements of purifying product for the sulphuretted hydrogen purifying are removed. The small amount of purifying mass in circulation in the process is always used over and over again.

g. The danger of explosion through the emptying and refilling of purifying tanks in the gas-works is completely removed.

For a coke works, the advantages, besides the economy in sulphuric acid and increase of ammonia yield, consist especially that all coke gases are completely freed from sulphuretted hydrogen, cyanogen and ammonia, and therefore can be used at once as lighting gas. Further, for coke works there is the very important hygienic advantage, which consists in the gases used for the burning in the ovens, being all free from sulphuretted hydrogen, and therefore all troubles from sulphurous acids are done away with.

NEUTRAL COKE

By J. R. Campbell
Scottdale, Pa.

SULPHUR IN NATURAL COKE

The tests show that about 33⅓% of the total sulphur in the coke made from washed Rend coal, was transformed to calcium sulphide during the coking process by the addition of 12½% limestone, perhaps, too small a percentage to have much metallurgical significance in blast furnace practice where the desideratum is to have the total sulphur in the form of calcium sulphide; also that the total sulphur in the neutral coke has actually increased about 10%, *i.e.*, a part of the otherwise volatile sulphur combined with the lime of the limestone during the coking process.

Furthermore, even if calcium sulphide was formed completely during the coking process, it would be extremely hazardous to assume that the calcium sulphide thus formed would pass unchanged through the blast furnace into the slag, in view of well-established claims that calcium sulphide is reduced by metallic oxides at low temperatures, or in other words, acts as a sulphurizing agent.

TEMPERATURE FOR NEUTRAL COKE

It is apparent that much higher temperatures, perhaps critical in the flues and oven walls, are required for the formation of calcium sulphide than ordinarily prevail during the coking process. At 2100 degrees F. in the coking mass, there is a 33⅓% transformation of sulphur, but there is no positive data to show that a 100% efficiency will be obtained at higher temperatures.

The free lime also is proportional to the temperature in the mass, though it appears to be easier to form a mono-silicate than calcium sulphide at the usual working temperatures. About 85% of the total lime in the neutral coke is in combination at 2100 degrees F.

It may also be pointed out that a part of the above deficiencies may be offset by finer pulverization of both coal and limestone.

PHYSICAL QUALITY OF NEUTRAL COKE

The physical quality of the calcite coke was much better than the usual coke in every instance, which is the one thing above all others that commends the Diehl-Faber process. The shatter test, porosity, etc., on neutral coke from washed coal from Rend mine No. 2, Illinois, was practically as good as the regular Klondike-Poca. coke, though it was smaller and still showed some spots.

By the use of the Diehl-Faber process, it is possible to transform an otherwise inferior grade of coke to a good blast furnace coke from a physical standpoint.

GAS AND AMMONIA YIELDS

There is an increase in the volume of gas and a decrease in the B.T.U. value from neutral coke, the former due perhaps to the high temperatures and the latter to high percentages of CO gas from limestone.

The total sulphate yield is as high from neutral coke with 12½% limestone as from regular coke, and 12½% higher when calculating back to coal actually charged. Neutral coke shows 35–40% more free ammonia and about 60% less fixed ammonia. The data on ammonia yields agrees with outside information which is that a 2% addition of lime increases the ammonia 10–15%.

COKING TIME FOR NEUTRAL COKE

The experiments indicate that the coking time for neutral coke will be increased about 33⅓% over the regular time, *i.e.*, if any real good is to be accomplished by the limestone addition.

The Effect of Calcium Chloride on the Ammonia Yield.

The results on bomb tests are as follows:

REND COAL TREATED WITH CALCIUM CHLORIDE

Ammonia as Sulphate	
(a) Free	0.48%
(b) Combined	1.50%
Total sulphate	37.2# per net ton

WASHED REND COAL WITHOUT CALCIUM CHLORIDE

Ammonia as Sulphate

(a) Free	1.33%
(b) Combined	0.45%
Total Sulphate	33.4 per net ton

The above chemical is used in the Campbell Chemical Washer. The total sulphate yield is increased 3.8 per net ton which means about 11 ¢ more credit for by-products.

While the combined ammonia is radically increased by the use of the calcium chloride, necessitating the use of more lime in the stills in the Koppers process, the available information is that there will not be any great interference with the present practice and that the advantages will more than offset the disadvantages.

The Effect of Iron on the Ammonia Yield

The following is the data prepared by the laboratory.

	Sulphate per net ton	
20% Rend —80% Poca.	22.4	
Rend with 10% Fe (filings)	16.0	or a decrease of 28.57%
Rend with 10% Flue Dust (57.64% Fe)	11.8	or a decrease of 47.32%
Rend with 10% Flue Dust (51.62% Fe)	17.6	or a decrease of 21.42%
Rend with 10% FeS_2 From Rend Coal	23.6	or an increase or 5.35%
Rend with 10% FeS_2 From Rend Coal	22.6	or an increase of .88%

There seems to be no question but what iron and certain iron compounds decrease the ammonia yield. The outside information we have available is to the effect that 3% of metallic oxide in the form of flue dust reduces the ammonia yield 8%.

In passing it may be remarked that the low practical yield of ammonia *versus* the theoretical yield may be due to iron piping around the by-product coke ovens which could be further insulated.

Cost of Neutral Coke

Figuring on a mixture of 100% washed Rend coal and 12½% limestone, the present cost above, based on an 18 hr. coking time and 80% yield, will be increased about 50%, due to increased cost of crushing the limestone, a 33⅓% increase in coking time and a 25% decrease in coke yield.

The details of most of the above conclusions can be found in the report which follows:

NEUTRAL COKE TEST AT GARY COKE PLANT

WASHED REND COAL USED IN TEST

Object of Test.—The purpose of this test was to investigate the claims of the Diehl-Faber process for making neutral coke, in which, by the introduction of limestone into the charge in certain proportions, it is claimed that the sulphur in the raw coal is transformed into calcium sulphide during the coking process, and that the physical quality of the resultant coke is improved.

Furthermore, we have endeavored to carry our investigation on into the effect the limestone will have upon certain by-products, *i.e.*, the gases and the ammonia yield, also the temperature conditions that must prevail to secure the desired results.

The details are now set forth from which certain conclusions may be drawn.

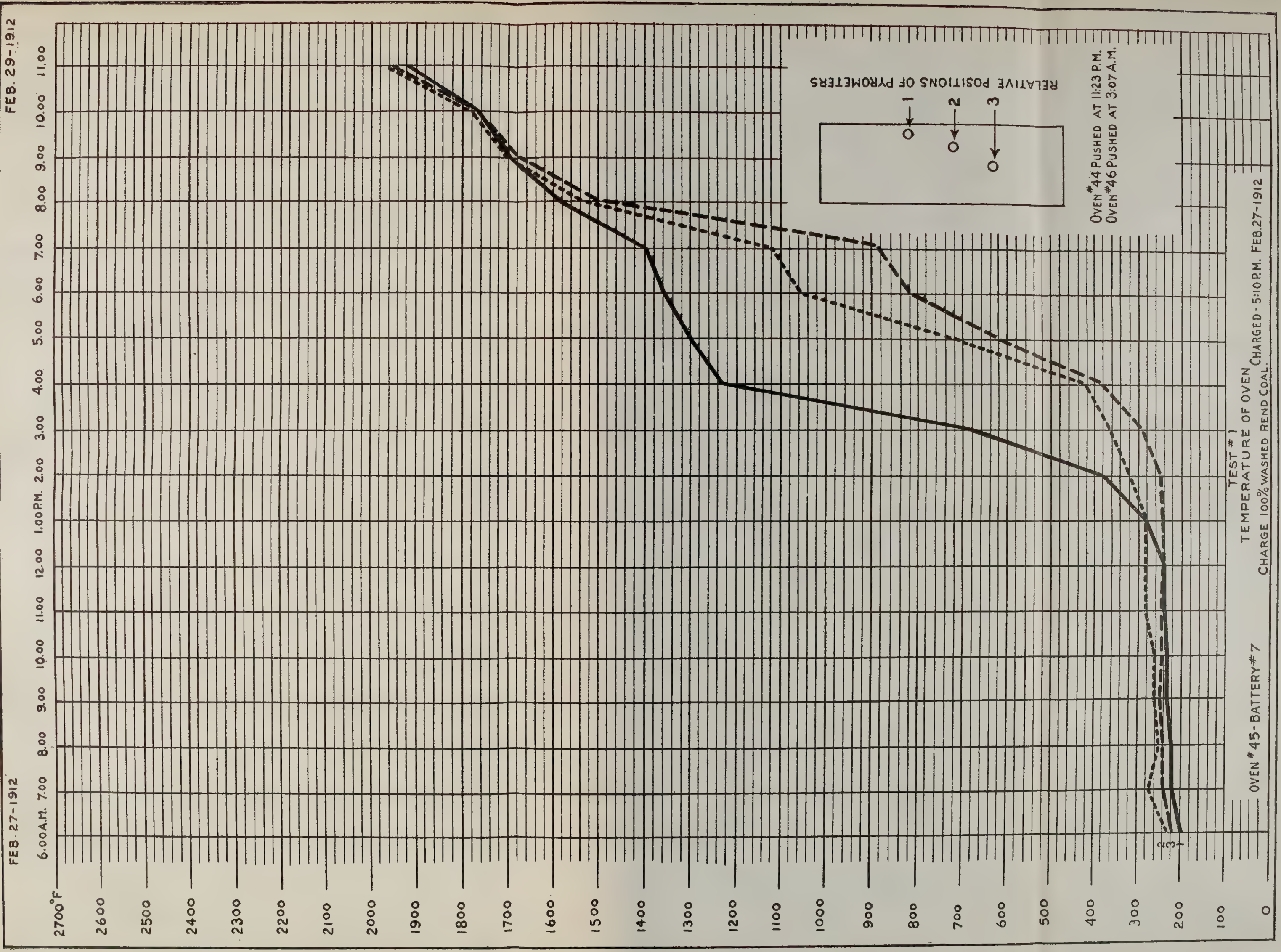
ANALYSES ASH IN WASHED REND COAL

SiO_2	Fe_2O_3	Al_2O_3	CaO	MgO	MnO	K_2O	Sul.	Month
48.44	15.58	22.52	5.10	1.14	.51	3.49	1.62	May 1911.
48.20	15.98	22.32	5.10	1.04	.59	3.55	1.64	June 1911.
45.76	17.58	22.12	6.20	1.08	.44	3.93	1.70	July 1911.
45.90	15.98	22.32	6.80	1.12	.49	4.14	1.75	Aug. 1911.
47.06	15.88	24.44	6.00	1.12	.56	3.92	1.34	Nov. 1911.
49.46	13.00	24.64	4.98	1.18	.49	4.67	1.27	Dec. 1911.
6)284.82	94.00	138.36	34.18	6.68	3.08	23.70	9.32	
47.48	15.67	23.06	5.69	1.11	.51	3.92	1.55	

WASHED REND COAL—DECEMBER, 1911

	Ash	Sulphur	Date
	9.03	2.46	Dec. 15th, 1911
	8.12	1.99	Dec. 16th, 1911
	9.06	2.18	Dec. 16th, 1911
	7.63	2.05	Dec. 17th, 1911
	8.73	2.18	Dec. 17th, 1911
	7.90	1.99	Dec. 18th, 1911
	6)50.47	12.85	
Average	8.41	2.14	

Note.—Two cars of washed Rend coal, were shipped for "neutral coke" test. Flux was calculated on the above average



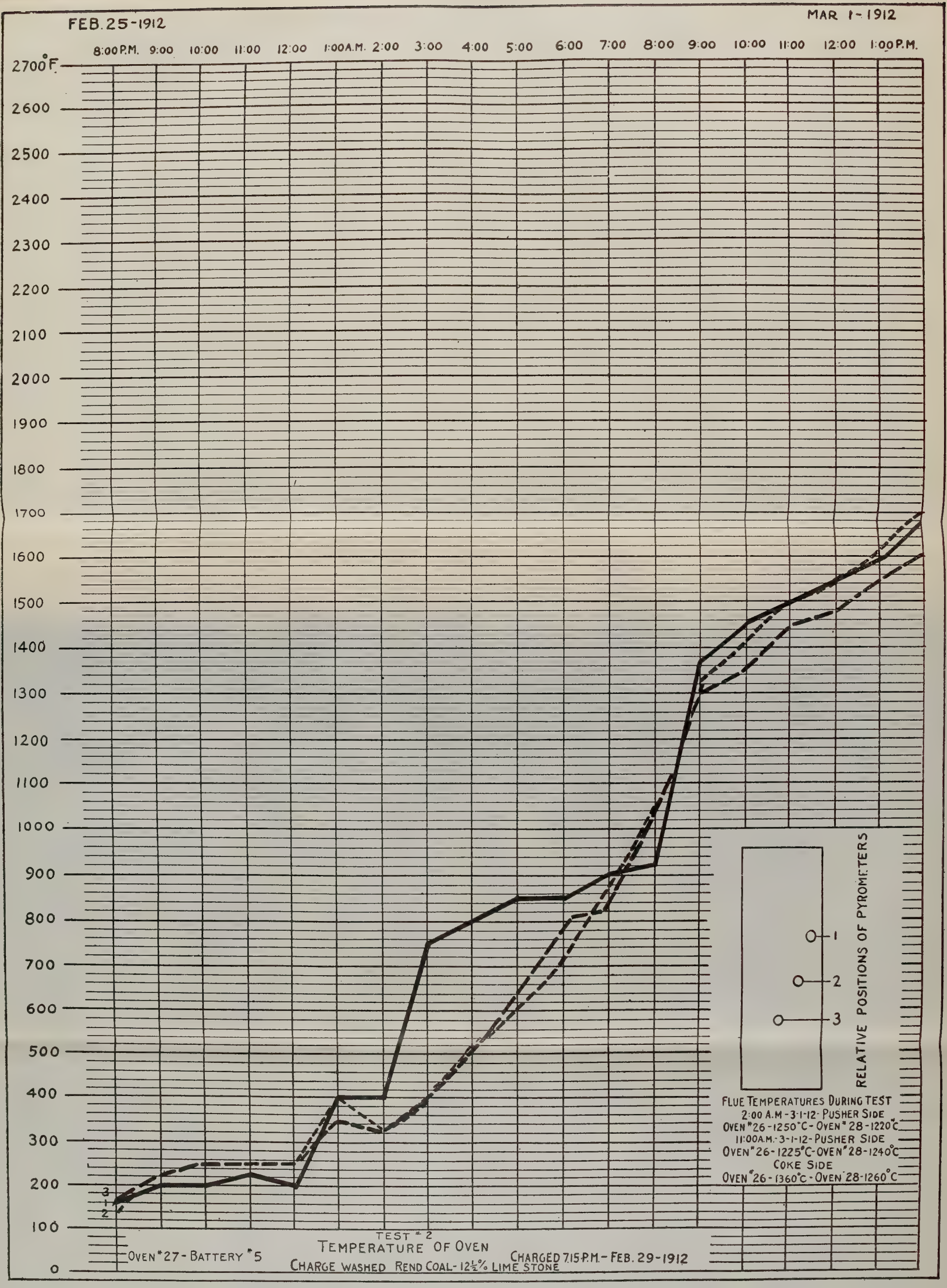
TEST #1

TEMPERATURE OF OVEN

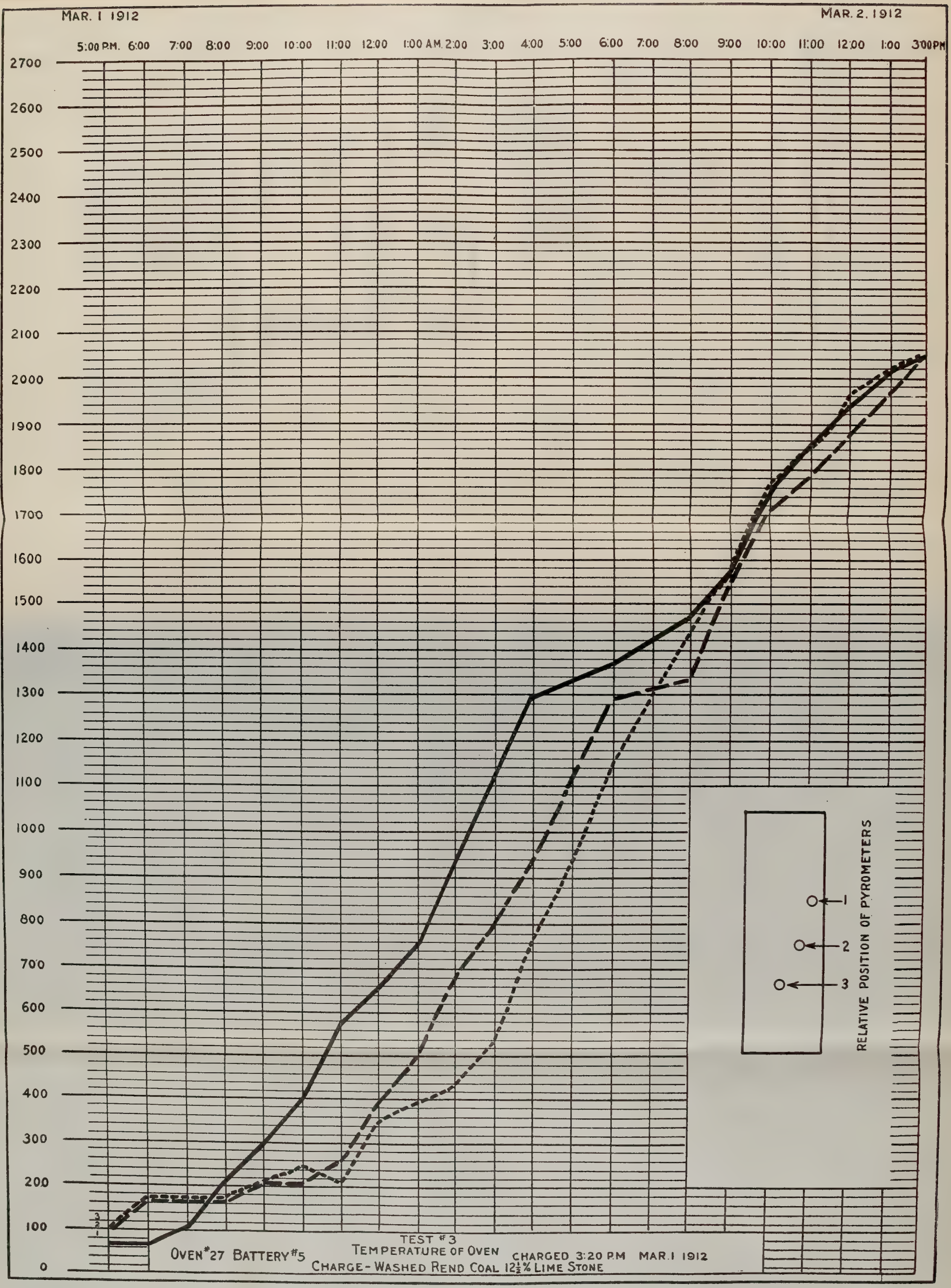
CHARGE 100% WASHED REND COAL. CHARGED - 5:10 P.M. FEB. 27-1912

OVEN #45-BATTERY #7

OVEN #44 PUSHED AT 11:23 P.M.
OVEN #46 PUSHED AT 3:07 A.M.



TEST #2
TEMPERATURE OF OVEN
OVEN #27 - BATTERY #5
CHARGE WASHED REND COAL - 12½% LIME STONE
CHARGED 7.15 P.M. - FEB. 29 - 1912

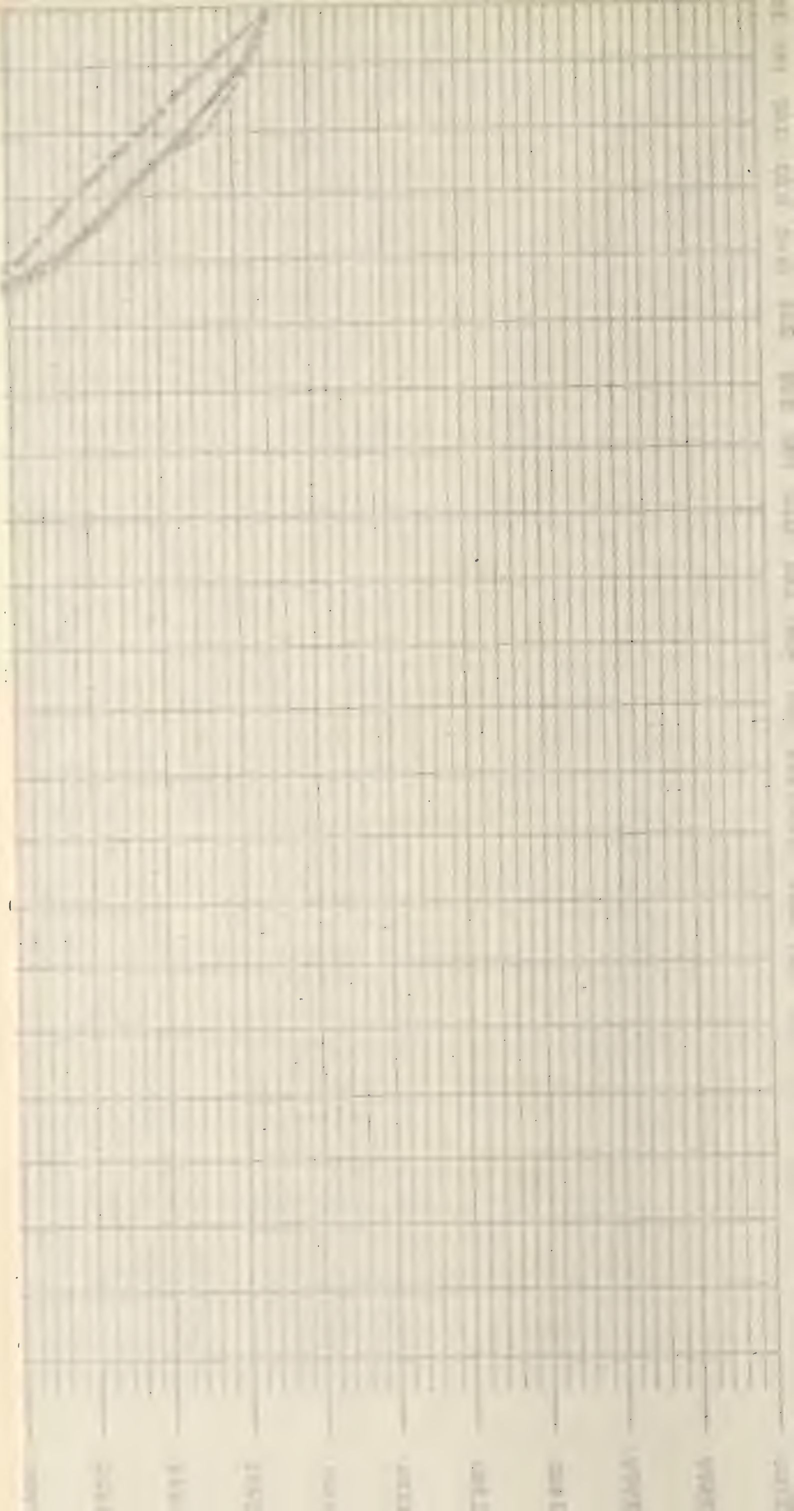


OVEN #27 BATTERY #5

TEST #3
TEMPERATURE OF OVEN

CHARGED 3:20 P.M. MAR. 1 1912

CHARGE - WASHED REND COAL 12½% LIME STONE



analyses of ash but carefully checked out with analyses of coal while test was in progress. These results are given elsewhere.

ANALYTICAL DATA ON WASHED REND COAL

	Proximate Analysis		Composition of Ash	
	Dry	Wet		
			SiO_2	47.85
V.M.	39.05	——	Fe_2O_3	15.84
F.C.	52.34	——	Al_2O_3	23.98
Ash	8.61	——	CaO	4.41
Sul.	2.09	——	MgO	1.66
H_2O		10.19	Sul.	1.11

FORMS OF SULPHUR IN COAL

Sulphur as Sulphides	1.17
Sulphur as Sulphates	.10
Other forms of Sulphur	.82
Total Iron	.93

SCREENING TEST

On ¼″	1.77	Thru	98.23
On ⅛″	9.29	Thru	90.71

LIMESTONE USED IN NEUTRAL COKE TEST
ANALYSIS LIMESTONE

SiO_2	Fe_2O_3	1.33
Al_2O_3		.63
CaO		54.12
MgO		1.25
CO_2, etc.		42.67

SCREENING TEST ON LIMESTONE

On 100 Mesh Screen	63.4%
On 80 Mesh Screen	58.2%
On 50 Mesh Screen	29.4%
On 40 Mesh Screen	21.6%
On 30 Mesh Screen	7.8%
On 20 Mesh Screen	.8%
On 10 Mesh Screen	Nothing

COKING TEST NO. 1

OVEN NO. 45. 100% WASHED REND COAL, BATTERY NO. 7

Weight of Coal Charged		24300	21821
H_2O in Coal	10.2%		
Weight of Coke Pushed		20600	13987
H_2O in Coke	32.1%		
Weight of breeze		580	383
H_2O in breeze	34.0%		
Net Yield Coke			64.2%
Time Charged	5:10 P.M. 2/27/12		
Time Pushed	12:30 P.M. 2/28/12		
Coking Time	19 hr. 20 min.		

PHYSICAL TESTS

Shatter Test (4 drops 2″ screen)	70.5%
Porosity	38.8%
Apparent sp. gr.	.900
Real sp. gr.	1.470

COKING TEST NO. 2

OVEN NO. 27 WASHED REND COAL AND 12½% LIMESTONE, BATTERY NO. 5

Weight of Mixture Charged		24600	22435
H_2O in mixture	8.8%		
Weight of Coke Pushed		15200	13376
H_2O in Coke	12.0%		
Weight of breeze		392	304
H_2O in breeze	22.4%		
Net Yield Coke			59.6%
Time Charged	7:80 P.M. 2/29/12		
Time Pushed	2:20 P.M. 3/1/12		
Coking Time	19 hr. 12 min.		

PHYSICAL TESTS

Shatter test	31.9%
Porosity	43.9%
Apparent Sp. Gr.	.934
Real Sp. Gr.	1.666

Coking Test No. 3

Oven No. 27. Washed Rend Coal and 12½% Limestone, Battery No. 5

Weight of Mixture Charged		24000	22080
H_2O in mixture	8.0%		
Weight of Coke Pushed		17400	14529
H_2O in Coke	16.5%		
Weight of Breeze		412	328
H_2O in Breeze	20.5%		
Net Yield Coke			65.8%

Time Charged	3:23 P.M. 3/1/12
Time Pushed	4:09 P.M. 3/2/12
Coking Time	24 hr. 46 min.

Physical Tests

Shatter test	38.0%
Porosity	45.2%
Apparent Sp. Gr.	.957
Real Sp. Gr.	1.748

Coking Test No. 4

Oven No. 70. Washed Coal and 18¾% Limestone Battery, No. 7

Time Charged	2:16 P.M. 3/4/12
Time Pushed	12:13 P.M. 3/5/12
Coking Time	21 hr. 57 min.

Physical Tests

Shatter Test	55.2%
Porosity	41.2%
Apparent Sp. Gr.	.969
Real Sp. Gr.	1.649

TEMPERATURES IN COKING MASS

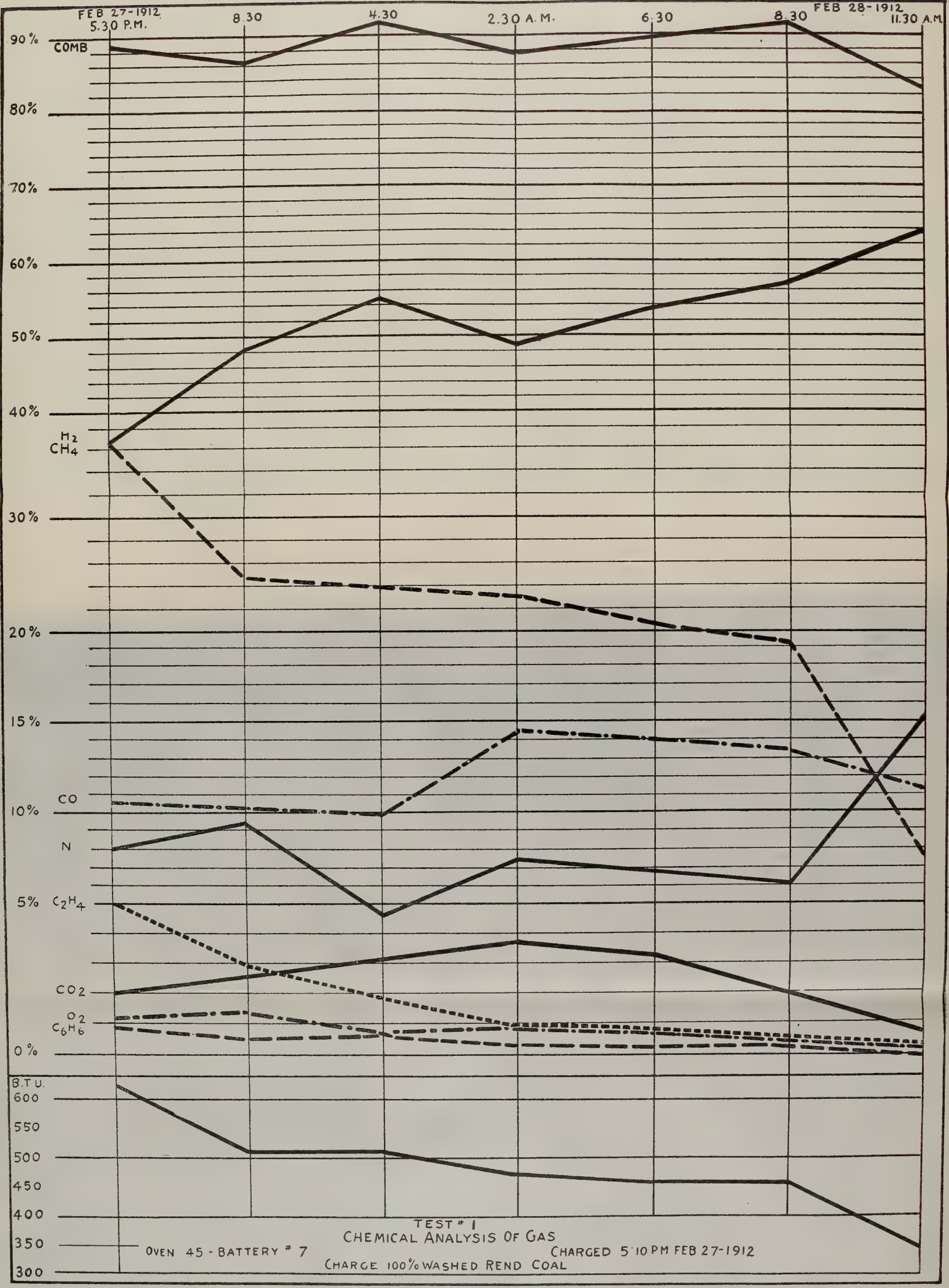
(Blue prints showing curves herewith)

Test No. 1

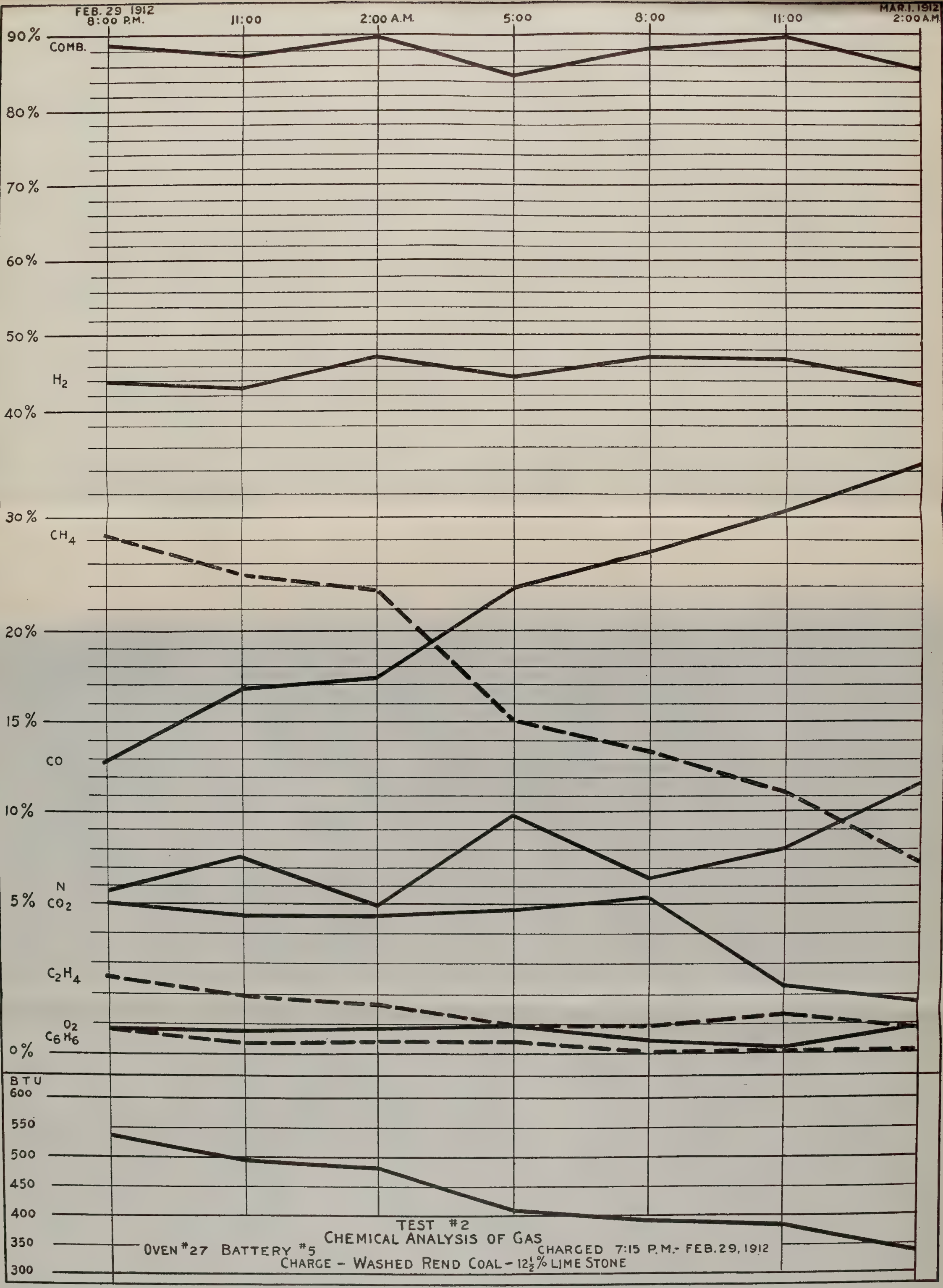
Time	Hole No. 1	Hole No. 2	Hole No. 3
5:30 P.M.	350	325	275
6:00 P.M.	200	225	200
6:30 P.M.	200	225	200
7:00 P.M.	200	270	250
8:00 P.M.	220	250	240
9:00 P.M.	230	260	250
10:00 P.M.	230	260	240
11:00 P.M.	240	280	240
12:00 M.	240	280	240
1:00 A.M.	280	290	250
2:00 A.M.	380	320	250
3.00 A.M.	690	360	290
4:00 A.M.	1220	420	380
5:00 A.M.	1300	720	620
6:00 A.M.	1360	1060	822
7:00 A.M.	1400	1120	880
8.00 A.M.	1580	1550	1510
9:00 A.M.	1700	1700	1680
10:00 A.M.	1770	1770	1770
11:00 A.M.	1920	1920	1950

Test No. 2

Time	Hole No. 1	Hole No. 2	Hole No. 3
8:00 P.M.	175	150	175
9:00 P.M.	200	225	225
10:00 P.M.	200	225	250
11:00 P.M.	225	225	250
12:00 M.	250	250	250
1:00 A.M.	400	400	350
2:00 A.M.	400	325	325
3:00 A.M.	750	400	400
4:00 A.M.	800	500	500
5:00 A.M.	850	600	650



TEST # 1
CHEMICAL ANALYSIS OF GAS
OVEN 45 - BATTERY # 7
CHARGED 5.10 PM FEB 27-1912
CHARGE 100% WASHED REND COAL

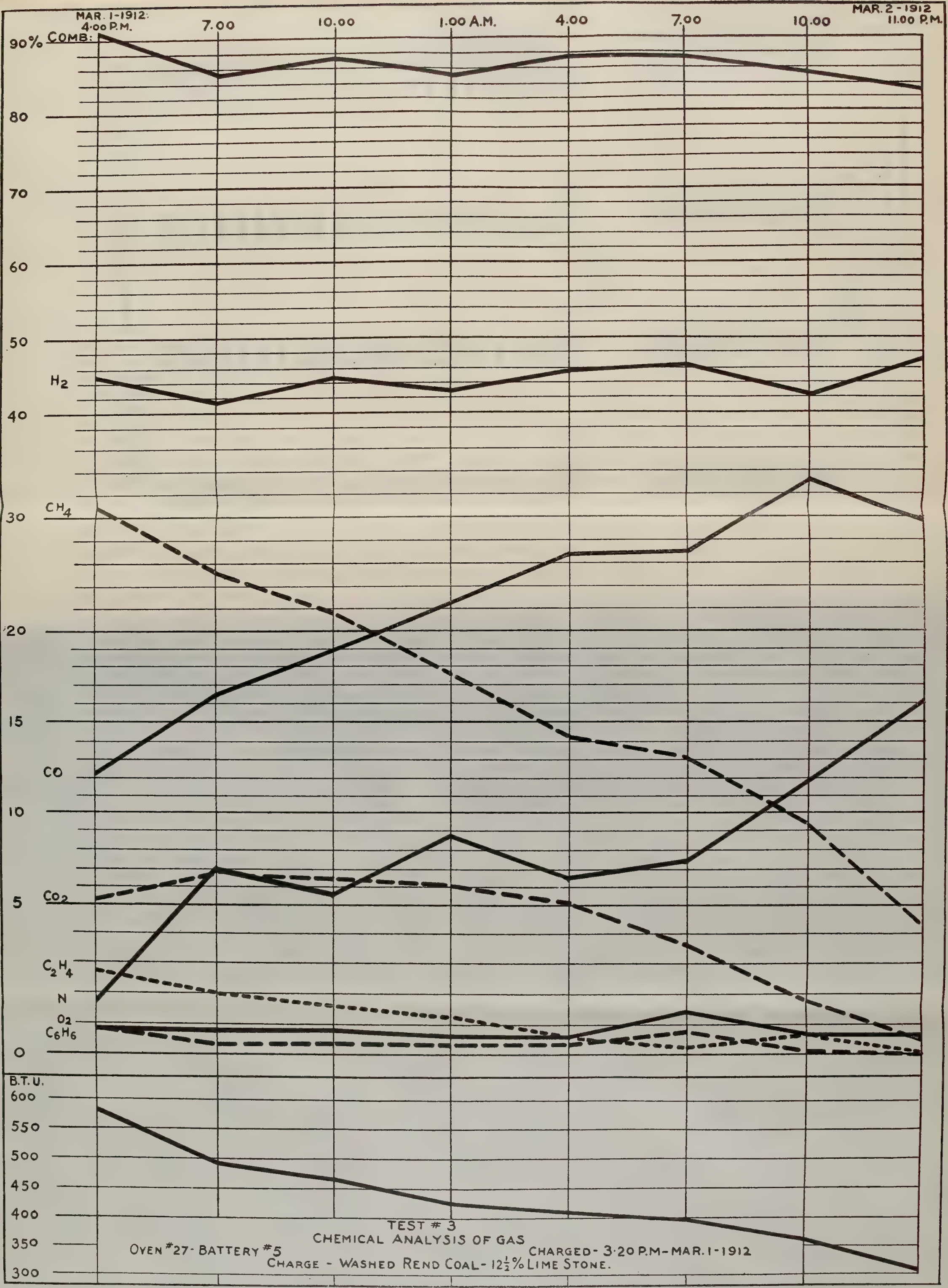


TEST #2

CHEMICAL ANALYSIS OF GAS

OVEN #27 BATTERY #5 CHARGED 7:15 P.M.- FEB. 29, 1912

CHARGE - WASHED REND COAL - 12½% LIME STONE



TEST # 3
CHEMICAL ANALYSIS OF GAS
OVEN #27 - BATTERY #5 CHARGED - 3·20 P.M. - MAR. 1-1912
CHARGE - WASHED REND COAL - 12½% LIME STONE.

1
1
1

1
1

8
9
10
11
12
1
2
3
4:
5:

Test No. 2—Continued

Time	Hole No. 1	Hole No. 2	Hole No. 3
6:00 A.M.	850	700	800
7:00 A.M.	900	880	825
8:00 A.M.	925	1025	1050
9:00 A.M.	1375	1320	1300
10:00 A.M.	1450	1410	1350
11:00 A.M.	1500	1500	1450
12.00 M.	1550	1545	1475
1:00 P.M.	1600	1610	1550
2:00 P.M.	1670	1700	1610

Test No. 3

Time	Hole No. 1	Hole No. 2	Hole No. 3
5:00 P.M.	68	104	104
6:00 P.M.	68	176	176
7:00 P.M.	104	176	176
8:00 P.M.	212	176	176
9:00 P.M.	294	212	212
10:00 P.M.	392	248	212
11:00 P.M.	572	212	248
12:00 M.	644	356	392
1:00 A.M.	752	392	500
2:00 A.M.	932	428	680
3:00 A.M.	1112	536	788
4:00 A.M.	1292	788	932
5:00 A.M.	1358	932	1112
6:00 A.M.	1394	1148	1292
8:00 A.M.	1472	1436	1358
9:00 A.M.	1580	1580	1544
10:00 A.M.	1760	1760	1706
11:00 A.M.	1868	1850	1780
12:00 M.	1940	1958	1886
1:00 P.M.	2016	2016	1960
3:00 P.M.	2050	2050	2050

Note.—The above readings expressed in degrees Fahrenheit.

CHEMICAL ANALYSES GASES

(Blue prints showing curves herewith)

Test No. 1—2/27/12

Gas Sampled at	5.30 P.M.	8.30 P.M.	11.30 P.M.	2.30 A.M.	5.30 A.M.	8.30 A.M.	11.30 A.M.
CO_2	2.00	2.60	3.20	3.80	3.40	2.00	0.60
C_6H_6	0.90	0.60	0.70	0.40	0.30	0.40	0.00
C_2H_4	5.00	3.00	1.90	1.00	0.90	0.70	0.40
O_2	1.20	1.40	0.80	1.00	0.80	0.60	0.40
CO	10.50	10.20	9.90	14.60	14.10	13.50	11.40
H_2	36.20	48.10	54.90	48.50	53.30	57.20	64.10
CH_4	36.20	24.70	24.00	23.20	20.50	19.50	7.50
N_2	8.00	9.40	4.60	7.50	6.70	6.10	15.40
Comb.	88.80	86.60	91.40	67.70	89.10	91.30	93.40
B.t.u	622.00	511.00	512.00	472.00	454.00	456.00	339.00

Test No. 2

Gas Sampled at	8.00 P.M.	11.00 P.M.	2.00 A.M.	5.00 A.M.	8.00 A.M.	11.00 A.M.	2.00 P.M.
CO_2	5.0	4.6	4.6	4.8	5.1	2.2	1.8
C_6H_6	.7	.4	.4	.4	0.0	0.0	0.0
C_2H_4	2.6	1.9	1.6	.8	0.8	1.3	.6
O_2	0.8	.7	.8	.8	0.4	.1	.7
CO	12.9	16.7	17.2	23.9	26.9	30.3	34.3
H_2	44.0	43.3	47.0	44.5	47.0	46.9	43.8
CH_4	28.4	24.9	23.5	15.0	13.4	11.2	7.2
N_2	5.6	7.5	4.9	9.8	6.4	8.0	11.6
Comb.	88.6	87.2	89.7	84.6	88.1	89.7	85.9
B.t.u.	542.00	494.00	488.00	403.00	390.00	386.00	337.00

Test No. 3

Gas Sampled at	4.00 P.M.	7.00 P.M.	10.00 P.M.	1.00 A.M.	4.00 A.M.	7.00 A.M.	10.00 A.M.	1.00 P.M.
CO_2	5.2	6.8	6.4	6.0	5.0	3.6	1.7	.5
C_6H_6	.8	.4	.4	.4	.4	.7	.1	.0
C_2H_4	2.8	2.0	1.6	1.2	.6	.3	.6	1.0
O_2	.8	.7	.7	.6	.4	1.4	.7	.6
CO	12.3	16.5	10.9	22.4	26.8	26.9	33.5	29.9
H_2	44.6	41.5	44.7	43.0	45.8	46.3	42.2	47.6
CH_4	30.7	25.2	21.8	17.6	14.3	13.2	9.4	4.4
N_2	2.8	6.9	5.5	8.8	6.7	7.6	11.8	16.0
Comb.	91.2	85.6	87.4	84.6	87.9	87.4	85.8	82.9
B.t.u.	582	492	469	426	403	392	355	313

ANALYTICAL DATA ON NEUTRAL COKE TEST

Test No. 1. 100 % Washed Rend Coal

	Proximate Analysis Coke		*Composition of Ash*	
	Dry	Wet		
V.M.	0.90		SiO_2	46.87
F.C.	85.51		Fe_2O_3	15.61
Ash	13.59		Al_2O_3	24.12
Sul.	1.62		CaO	4.21
H_2O		32.1	Mgo	1.41
			Sul.	.86

Sulphur as CaS	trace
Sulphur as $CaSO_4$	.22

Test No. 2. Washed Rend Coal and 12½% Limestone

	Proximate Analysis Coke		*Composition of Ash*	
	Dry	Wet		
V.M.	4.66		SiO_2	24.06
F.C.	69.60		Fe_2O_3	11.54
Ash	25.74		Al_2O_3	9.93
Sul.	1.37		CaO	48.00
H_2O		12.00	MgO	1.15
			Sul.	.61

Sulphur as CaS	.25%
Sulphur as $CaSO^4$	.27%
Free CaO	2.85%

Test No. 3. Washed Rend Coal and 12½% Limestone

	Proximate Analysis Coke			
	Dry	Wet		
V.M.	2.34			
F.C.	72.70		Sulphur as CaS	.53%
Ash	24.96		Sulphur as $CaSO_4$	.33%
Sul.	1.76		Free CaO	1.65%
H_2O		16.5		

Test No. 4. Washed Rend Coal and $18\frac{3}{4}\%$ *Limestone*
Proximate Analysis Coke

	Dry	Wet		
V.M.	7.54			
F.C.	64.62		Sulphur as CaS	
Ash	27.84		Sulphur as $CaSO_4$	
Sul.	1.61		Free CaO	5.10%
H_2O		19.2		

We Conclude

1. That the temperature of the coking mass is insufficient for the complete transformation of S in the coal to CaS in the coke during the coking process, and that such temperature must be over 2100° F. perhaps near 2500° F. as evidenced by the foregoing data.

2. That the temperature is insufficient for the complete formation of lime silicates, as evidenced by the free CaO in Tests 2 and 3.

3. That it is not practicable to use more limestone than will combine with the ash of the coal to form a mono-silicate, as evidenced by Test No. 4.

4. That the physical quality of the coke is improved by the limestone additions if too much free CaO does not remain in the coke and if such additions are for the formation of a mono-silicate, as evidenced by the physical tests of the coke and its physical appearance, provided, always that high temperatures prevail at or near the close of the coking process.

5. That the decomposition of the limestone and the action of the resultant CO_4 on the incandescent coke, forms high percentages of CO gas, which lowers the B.t.u. value of the whole gas about 10%, as evidenced by the gas analyses.

6. That it follows naturally from 5, the percentage of yield from neutral coke is probably lower than the R/M coke, Tests No. 2 and No. 3 showing between 1% and 2% lower yield on an average than Test No. 1.

7. That the coking time will be extended by the introduction of limestone into the charge, if any beneficial results are to be expected, as evidenced by Tests No. 1 and No. 3.

8. That the total NH_3 yield will be increased quite materially as evidenced by bomb tests, the details of which will be given in a separate report.

9. That to attain sufficiently high temperature in the coking mass for the desired chemical reaction and permanent physical qualities in the coke, will necessitate maintaining the flues at critical temperatures, say: 2700° to 3000° F.

10. That the Diehl-Faber process has but little practical value from a blast furnace standpoint, unless all the sulphur is converted to CaS during the coking process, or unless a silicate is formed, for the reasons that S other than CaS will enter the iron readily and that the free CaO will cause the coke to crumble upon exposure to the atmosphere.

NEUTRAL COKE

80% Pocahontas 20% Washed Rend and 11% Limestone

Analysis of Ash

	SiO_2	Fe_2O_3	Al_2O_3	CaO	MgO	K_2O
Pocahontas 80%	47.46	13.90	31.90	2.52	1.99	1.89
Washed Rend 20%	47.47	15.67	23.06	5.69	1.11	3.92
Average	47.46	14.25	30.13	3.15	1.81	2.29

PROXIMATE ANALYSES COAL

	20% Washed Rend	80% R/M Pocahontas	100% Mixture
V.M.	39.05	19.09	23.08
F.C.	52.34	73.07	68.92
Ash	8.61	7.84	8.00
Sul.	2.09	.70	.98

Note.—Mixture was based on the above ash analyses, and the pulverization of both coal and limestone was same as in test using washed Rend coal.

COKING TEST NO. 1

Oven No. 47. 80% Pocahontas and 20% Washed Rend,
Battery No. 5

Time Charged	4:38 P.M. 3/7/12
Time Pushed	9:16 A.M. 3/8/12
Coking Time	16 hr. 38 min.

Temperature as pushed.

Hole No. 1 (Hoskins)		1814° F.
Hole No. 2 (Hoskins)		1814° F.
Hole No. 3 (Hoskins)		1830° F.
On the Coke Mass	(Warmer)	1938° F.
On the oven walls	(Warmer)	1992° F.
Flue on No. 46 (pusher side)	(Warmer)	2370° F.
Flue on No. 48 (pusher side)	(Warmer)	2300° F.

PHYSICAL TESTS

Shatter Test	30.5%
Porosity	45.2%
Apparent Sp. Gr.	.972
Real Sp. Gr.	1.774

CHEMICAL ANALYSES

	Dry	Wet		
V.M.	1.14			
F.C.	87.41		Sulphur as CaS	nil
Ash	11.45		Sulphur as $CaSO_4$	trace
Sul.	.61		Free CaO	nil
H_9O		7.00		

COKING TEST NO. 2

Oven No. 7. 80% Pocahontas, 20% Washed Rend, and 11¾%
Limestone, Battery No. 7

Time Charged	1:27 P.M. 3/8/12
Time Pushed	1:05 P.M. 3/9/12
Coking Time	23 hr. 38 min.

Temperatures as pushed.

Hole No. 1	(Warmer)	Not taken
Hole No. 2	(Warmer)	1953° F.
Hole No. 3	(Warmer)	1895° F.
On Coke Mass	(Warmer)	2010° F.
On Oven Walls	(Warmer)	2156° F.
Top of Coke		
Hole No. 1	(Warmer)	2005° F.
Hole No. 2	(Warmer)	2055° F.
Hole No. 3	(Warmer)	2093° F.
Flues on pusher side		
Oven No. 6	(Warmer)	2354° F.
Oven No. 8	(Warmer)	2354° F.
Flues in middle		
Oven No. 6	(Warmer)	2397° F.
Oven No. 8	(Warmer)	2426° F.

PHYSICAL TESTS

Shatter Test	19.0%
Porosity	42.6%
Apparent Sp. Gr.	1.121
Real Sp. Gr.	1.953

CHEMICAL ANALYSES

	Dry	Wet		
V.M.	1.40		Sulphur as CaS	.097%
F.C.	77.85		Sulphur as $CaSO_4$	.15%
Ash	20.75		Free CaO	1.40%
Sul.	.69			
H_2O		2.60		

Conclusion

The coke from the 80% Pocahontas—20% washed Rend and 11$\frac{3}{4}$% limestone was much larger and showed fewer spots than the "blank" charge, and the transformation of S to CaS was a very small percentage — .097%, as evidenced by the physical and chemical data.

It is interesting to note that a fusible ash in the coke of all limestone charges was produced, as, upon ashing the coke in analysis at high temperatures, it was fused completely.

Bomb Tests

Washed Rend Coal and 12½ Limestone

Test No. 1

Kind of bomb	Large Iron Bomb
Size of Charge	453.6 grams
Temperature of Muffle	
(a) at beginning	2240° F.
(b) at end	2450° F.
Duration of test	1 hr. 5 min.
Yields	
(a) Coke	lost
(b) Sulphate	
1. Free	20.36
2. Combined	2.03
(c) Gas	11,000 cu. ft. at 75° F.
(d) Tar	not done

Chemical Analyses

V.M.	2.30	Sulphur as CaS	.28%
F.C.	68.79	Sulphur as $CaSO_4$	.21%
Ash	28.91	Free CaO	1.20%
Sul.	1.68		

Note.—The iron bomb melted at end of test. The object of this test was to show effect of high temperature upon the formation of CaS, but it is evident that either the temperature of the muffle was not attained in the bomb, or, if attained, not for a long enough time to produce complete reaction, as evidenced by the CaS—.28.%

Bomb Test No. 2

Washed Rend Coal

Kind of Bomb	Small Iron Bomb
Size of Charge	30 grams
Temperature of Muffle	
(a) at beginning	1652° F.
(b) at end	1652° F.
Duration of test	45 min.

Yields

(A) Coke	59.1%
(B) Sulphate	
1. Free	16.68
2. Combined	5.70
(C) Gas	6653 cu. ft.
(D) Tar	not done

CONCLUSION ON BOMB TESTS

1. That if "neutral coke" were made from washed Rend coal the gas and sulphate yields would be increased, per ton of actual coal charged, due to well understood chemical reactions.

2. That the percentage of combined NH_3 is radically decreased by the limestone additions, as evidenced by the data at hand.

3. That, while the volume of gas is increased by the limestone additions, the B.t.u. value is decreased; consequently, there will be a smaller percentage of *available* gas.

UNIFORM OIL INSPECTION FROM A LEGAL POINT OF VIEW

By C. D. Chamberlain
Cleveland, Ohio

The question was once asked of a famous jurist by a student in his office as to what is the first essential in winning a law suit. The answer was, "Facts." The first question your attorney will ask you when you bring your case to his attention to engage his services in your behalf is, "What is your proof?" He must know all the facts, and so in discussing any question the first inquiry should be for all the facts pertinent and material to the question to be examined. So in this question, what are the facts?

Petroleum is the substance dealt with. What is it? Webster says:

"Rock oil, mineral oil, or natural oil, a dark brown or greenish inflammable liquid, which at certain points exists in the upper strata of the earth, whence it is pumped or forced by pressure of the gas attending it.

"Petroleum consists of a complex mixture of various hydrocarbons, and varies much in appearance, composition and properties.

"Petroleum is refined by fractional distillation, yielding successively volatile products, kerosene, lubricating oils and paraffine."

Webster gives the volatile products in their order of volatility from Pennsylvania crude petroleum as:

"Products	Sp. Gr.	Boiling point	Uses
Cymogene	110° B.	32° F.	Refrigerant.
Rhygolene	100° B.	65° F.	Anæsthetic
Petroleum Ether	81° B.	158° F.	Carburetant
Gasoline	73° B.	194° F.	Solvent; fuel; illuminant.
Naphtha	70° B.	230° F.	Solvent; fuel; illuminant.
Ligroine	62° B.	248° F.	Solvent; illuminant.
Benzine	57° B.	300° F.	Solvent."

Cymogene is a gas except at low temperatures. The others are volatile fluids, and in the ordinary process of refining the first two are not produced.

Following in the order of distillation is kerosene, below which the products are not required except in a few states to meet any tests; hence need not be considered.

Commercially the volatile fluids are of from two to four times the value of kerosene, and are carefully extracted from it by a second distillation by steam.

Technically none of the products of petroleum are explosive, and are dangerous only because of their volatility and inflammability.

When hydro-carbon vapor is mingled with air in the right proportions to form a critical mixture, such mixture will, if ignited, explode with much force. The critical range is from 5 to 15% of vapor.

USES

The products of petroleum are employed in a multitude of uses too numerous in a brief address to mention, but so far as put to any test by legal requirements in this country the chief are for illumination, heat, power and as solvents, and the entire series of such products are for those purposes.

CHARACTERISTICS

Petroleum is produced from several strata in New York, Pennsylvania, Ohio, West Virginia, Tennessee and Kentucky on the western slope of the Appalachian Range, the axis of production corresponding with that of the range. This is called "Pennsylvania crude" and is about 44 degrees B., its base being paraffine.

The Pennsylvania field is the oldest,—discovered in 1859. The next chronologically is the California field, of a heavy asphaltum base and about 24 degrees B. The Trenton Rock field of Ohio and Indiana comes next in point of time, and is of paraffine base of about 36 degrees B. impregnated with sulphur. The Kansas-Oklahoma or Mid-Continent field follows, of a mixed asphaltum and paraffine base of about 34 degrees B. At about the same time the costal production of Texas came in, of a heavy asphaltum base similar to the California crude and of about the same gravity B. which extended to Louisiana where the oil was of lighter gravity and contains paraffine. The next and last important field is that of Illinois, of a mixed base of paraffine and asphaltum.

The products from the crudes of all fields are similar in their main characteristics and differ only in quantities of each produced and in density and purity, or at least so nearly that, from a legal consideration, they may be regarded the same.

BASIS OF INSPECTION LAWS

The right to require that products of petroleum be subjected to certain tests before being offered to the public for use rests upon the maxim "*Salus populi suprema est lex.*" The power to provide and enforce this law is one of the powers reserved to the states, except in the proper regulation of interstate commerce by the Federal Government.

In *United States* vs. *Dewitt, 9 Wall, 41*, the Supreme Court say:

"By the Internal Revenue Act of March 2, 1867, 14 Statutes at Large 484, a penalty was imposed upon any person who should mix for sale naphtha and illuminating oils, or who knowingly sells or keeps for sale such mixture, or who should sell or offer for sale oil made from petroleum for illuminating purposes, inflammable at less temperature or fire test than 110 degrees Fahrenheit.

"We held that to be simply a police regulation, relating exclusively to the internal trade of the States; although emanating from Congress, that it could have by its own force no constitutional operation within State limits, and was without effect, except where the legislative authority of Congress excluded, territorially, all State legislation, as for example, the District of Columbia."

In *Patterson* vs. *Kentucky, 97 U. S. 501*, the same Court say:

"By the settled doctrine of this Court the police power [of the States] extends, at least, to the protection of the lives, the health and the property of the community against the injurious exercise by any citizen of his own rights. State legislation strictly and legitimately for police purposes, does not, in the sense of the Constitution, necessarily intrench upon any authority which has been confided expressly or by necessary implication to the National Government.

"The Kentucky statute under examination manifestly belongs to that class of legislation. It is in the best sense, a mere police regulation deemed essential for the protection of the lives and property of its citizens. It expresses in the most solemn form the deliberate judgment of the State that burning fluids which ignite and permanently burn at less than a prescribed temperature are unsafe for illuminating purposes. Whether the policy thus pursued by the State is wise or unwise, is not in the province of the national authorities to determine. That belongs to each State under its own sense of duty, and in view of its own constitution. Its action in those respects, is beyond the corrective power of this Court."

The above opinions from the highest judicial authority clearly place the necessary supervision of what regulations are to be made in the sale and use of petroleum products within the police power of the states. This power restrains and regulates, for the promotion of the public welfare, the natural or common liberty of the citizen in the use of his personal faculties and of his property. It rests upon the legal maxim, "*Sic utere tuo ut alianum laedas.*" The power is sovereign and has been spoken of as infinite supremacy; but the exercise of this power, that is, the regulations prescribed by legislative enactment, must be reasonable.

In the case of *W. M. & P. R. R. Co.* vs. *Jacobson, 179 U. S. 287*, the Supreme Court say:

"A statute or a regulation provided for therein, is frequently valid, or the reverse, according as the fact may be, whether it is a reasonable or an unreasonable exercise of legislative power over the subject matter involved, and in many cases questions of degree are the controlling ones by which to determine the validity or the reverse of legislative action."

And in *Davidson* vs. *New Orleans, 96 U. S. 97:*

"But it does not follow that every act of legislation is due process or the law of the land; and arbitrary statute is neither."

"Freund on Police Powers" at section 148 says:

"The peculiar difficulty of safety and health legislation is that the possible causes of injury to persons and property are extremely numerous and practically ubiquitous, that there is hardly any industry in which they may not be found if sought for, and that while the danger is often slight and remote the measure devised to combat it may profoundly affect economic interests, favoring one set of interests and prejudicing another.

"The questions which present themselves in the examination of a safety or health measure are: Does a danger exist? Is it of sufficient magnitude? Does the proposed measure tend to remove it? *Does it concern the public?* Is the restraint or requirement in proportion to the danger? Is it possible to secure the object sought without impairing essential rights and principles? Does the choice of a particular measure show that some other interest than safety or health was the actual motive of the legislation?"

From the above, then, it would seem that if there is danger more than ordinarily attaching to other commodities of sale and use that there should be inspection. If not, then, inspection is a useless burden.

From the facts stated as a premise, it would seem that no danger exists in the sale and use of petroleum and its products calling for the regulations which exist in many states. Apply the test laid down by Freund, above quoted, each question would be answered in the negative. Danger does not exist of sufficient magnitude which the regulations tend to remove. In other words, no test applied to gasoline reduces its tendency to vaporize and its ease of ignition. All users of it know these characteristics, and the substance is bought and used because of them.

The tests usually prescribed for gasoline are the gravity test by Beaume scale, since the flash test is at or below 0. Obviously neither test tends to lessen danger, and neither gives notice to the public of any latent danger in the use thereof.

So of illuminating oil. The test of specific gravity is no warning to the public of any latent danger that would otherwise be unknown, and the usual fire or flash tests prescribed by the various statutes only tell the judgment of the legislature of each state what the point of safety by that test is when used for illumination, and are no index of absolute safety.

While there was in the early period of refining a reason for requiring an oil to stand a test that made it safe for use in illumination, such reason no longer exists. Commercial values have changed. Gasoline is worth three times illuminating oil. Refiners carefully extract, by modern methods of refining, every available fraction of the volatile matter. No refined illuminating oil is now made or offered for sale that is not above the test of safety as measured by the test for export, and, as a test for safety is all that the police power can properly prescribe, no exigency remains for it to provide against that a test will accomplish.

The only uses to which gasoline and illuminating oil or refined petroleum can be put are for the production of light, heat, power and mechanical use, and they are never used as foods or for pharmaceutical purposes; consequently, the police power, if called into action, could only be for the safety of users.

We can readily understand, however, from the known facts that practically the exertion of police power in this direction by prescribing any tests accomplishes nothing.

PRESENT CONDITIONS

From the standpoint of legislation, no subject is more diverse and chaotic than the laws of the different states on this. In the matter of test of illuminating oil for the flask point, we find that ten states have no oil inspection laws: Alabama, Arizona, California, Florida, Mississippi, Nevada, Oregon, South Carolina, Texas, Virginia and West Virginia.

Twenty-two States use the Tagliabue tester: Louisiana, 125°; Arkansas, Illinois, Indiana, Missouri, Tennessee, 120°; Oklahoma, 115°; Connecticut, Michigan, 110°; Wisconsin, 105°; Kentucky and New Hampshire, 100°; New Mexico, 95°; Idaho, Maine, Minnesota, Washington, 90°; Delaware 85°; Maryland, Massachusetts, Pennsylvania and Rhode Island, 80°.

Six States use Elliott's tester: Georgia, New York, North Carolina, 129°; Iowa, South Dakota and Wyoming, 134°.

Six States use Foster's tester: Michigan, Ohio, Montana, 110°; Nebraska, 102°; Kansas, 100°; North Dakota, 95°.

Three States have a test equal to 134° Flash Tagliabue,
Three States have a test equal to 129° Flash Tagliabue,
One State has a test equal to 125° Flash Tagliabue,
Five States have a test equal to 120° Flash Tagliabue,
One State has a test equal to 115° Flash Tagliabue,
Four States have a test equal to 110° Flash Tagliabue,

One State has a test equal to 105° Flash Tagliabue,
One State has a test equal to 102° Flash Tagliabue,
Five States have a test equal to 100° Flash Tagliabua,
Two States have a test equal to 95° Flash Tagliabue,
Four States have a test equal to 90° Flash Tagliabue,
One State has a test equal to 85° Flash Tagliabue,
Four States have a test equal to 80° Flash Tagliabue,
All have an average of 106° Flash Tagliabue.

In foreign countries a much greater uniformity prevails. Their tests are:

Great Britain — 73° Flash Test Abel, equal to 100° Flash,
Australia — 70° Flash Test Abel-Pensky, equal to 97° Flash,
New Zealand — 100° Flash Test Tagliabue, equal to 100° Flash,
Barbadoes — 83° Flash Test Abel, equal to 110° Flash,
Bermuda — 73° Flash Test Abel, equal to 100° Flash,
Br. Honduras — 100° Flash Test Tagliabue, equal to 100° Flash,
Canada — 85° Flash Test Abel, equal to 112° Flash,
Ceylon — 73° Flash Test Abel, equal to 100° Flash,
Denmark — 73° Flash Test Abel, equal to 100° Flash
France — 95° Flash Test Granier, equal to 90° Flash,
Germany — 69.8° Flash Test Abel-Pensky, equal to 96.8° Flash,
Holland — 69.8° Flash Test Abel-Pensky, equal to 96.8° Flash,
India — 73° Flash Test Abel-Pensky, equal to 100° Flash,
Italy — 75° Flash Test Abel-Pensky, equal to 102° Flash,
Japan — 86° Flash Test Abel-Pensky, equal to 113° Flash,
Norway — 71.6° Flash Test Abel-Pensky, equal to 98.6° Flash,
Russia — 82.4° Flash Test Abel-Pensky, equal to 109.4° Flash,
Spain — No test required,
Sweden — 71.6° Flash Test Abel-Pensky, equal to 98.6° Flash.
Average test of all foreign countries — 10½° Flash.

No uniformity exists in the instruments prescribed or the use of them, no uniformity in the execution and administration of the laws, and none in the charges or fees for inspection, so that, a refiner must consult the statutes of each state before making his oil ready for shipment, in fact, must manufacture each lot of oil to fit the requirements of the particular state at a considerable economic loss both in manufacturing and selling.

It is true that many of the laws would not stand the scrutiny of the courts when the facts were properly and intelligently brought to their notice in a judicial way, but the refiner cannot afford to challenge the law of each State by expensive litigation, with the legal presumption in favor of the statute no matter how arbitrary or unreasonable.

LEGAL PRECEDENTS

Probably the burden of contesting the validity of the laws governing the sale and use of petroleum products accounts for the few decisions that have been handed down and the fact that many of them are adverse to the facts well known to those engaged in the oil business. Certainly they are not sufficient in number or uniformity to invoke the rule of *stare decisis*. The tendency of such decisions as have been made probably from insufficient knowledge is even more alarming to the seller of oil than the exactions of statutes, and points to the ultimate liability of insurer by all sellers of oil. For instance, in *State* vs. *Boylan, 79 Conn. 463,* the court held:

"That inasmuch as the statute did not prescribe what instrument or method should be used in determining the point of inflammability, the law would require the use of such reasonably accessible instruments and methods as would most accurately ascertain that point; leaving the determination of what particular instrument or method was the most accurate and reliable, in case of a conflict upon that subject, to the decision of the jury."

Every one knows the tendency of the ordinary jury in a damage case, especially where a casualty is the outcome of the accident. Is the seller at the mercy of the jury in the State of Connecticut every time he sells oil there? That is the rule of law. The inspection or the law under which the inspector acts is no defense in a damage case, if it be the rule of law in Iowa, as held in *Hatcher* vs. *Dunn, 71 N. W. 343:*

"Under Acts 20 General Assembly c. 185 sec. 11, providing that if any inspector of illuminating oil shall falsely brand the same or be guilty of fraud or culpable negligence, he shall be deemed guilty of a misdemeanor and punished therefor, an inspecting officer who uses instruments approved and furnished by the State Board of Health, and which he has reason to believe are in bad order, if he uses them with due care, and correctly brands the oil inspected according to the results shown by the test, is not liable for any error which may have occurred."

Also in *Hourigan* vs. *Nowall, 110 Mass. 470:*

" In an action under the Statute of 1869 c. 152, to recover for injuries caused by the explosion of illuminating oil, which had been sold by defendant, and did not conform to the standard fixed by the statute, it is no defence either that the defendant was ignorant that it did not so conform, or that an authorized inspector has authorized that it did; and if the person injured was using such care as would have been proper had the oil conformed to that standard, the jury will be warranted in finding that she was in the exercise of due care."

The above decisions come dangerously near to being within the doctrine of *res ipsa loquitur.* The seller of illuminating oil may feel that he only guarantees his product as safe for the purposes for which it is sold, *viz.:* for illumination, and that if the purchaser uses it for kindling a fire, or accelerating a fire already burning, that he has no liability for damages resulting from such use, yet, in *Waters-Pierce Oil Co.* vs. *Deselms, 212 U. S. 158,* the court say:

" A court may judicially notice the existence in the community of a general custom to use oil in kindling fires. Submitting to the jury the question of the existence of a custom or usage to use coal oil in kindling fires is not prejudicial error, where the custom in the community is so universal as to justify the court in taking judicial notice of its existence."

This in spite of the almost proverbial negligence of the use of oil in kindling fires. Other cases might be cited to the same intent, but enough has been noticed to show the necessity for an intelligent study of the question, a correct and scientific analysis of what is proper and justifiable legislation, and what and who should fall within such regulation.

Oil and gasoline will not explode. When circumstances arise from which the vapor from either mingles with the atmosphere to a critical point, a severe explosion may follow ignition. The province of police power, then, is to make such regulations as will prevent such circumstances, and these can not be accomplished by any tests. The red can for gasoline, preventing its being mistaken for oil, is a proper regulation; forbidding the use of either except at one's own risk for kindling fires, is a proper police regulation; and a recognition of negligence *per se* by such misuse of carbon oil by judicial notice, will tend to lessen casualties and bring to the attention of the public that such danger is assumed at the peril of the user with no redress upon the seller.

This, society can not do better than to clarify the confusion of facts for use in judicial determination, and so relieve a necessary and useful business of an entanglement of embarrassments.

The most rational legislation that has been adopted is found in the Act of the Pennsylvania Legislature of 1911. It makes it the duty of the Manufacturer to test for safety and prescribes the test and methods of testing, and provides a penalty for failure to comply. The text of the statute is:

"LAWS OF PENNSYLVANIA, SESSION OF 1911,
(Page 869)

AN ACT

"*To provide a standard of tests for illuminating oils, to provide a penalty for violating the provisions of this act, and repealing former acts.*

"Section 1. Be it enacted, &c., That no refined petroleum oil for illuminating purposes shall be manufactured, sold or offered for sale by any person, firm, or corporation, within the Commonwealth of Pennsylvania having a fire test lower than 110 degrees, Tabliabue's open cup, according to the following formula: Fill the water-bath of the apparatus with water of a temperature under 80 degrees Fahrenheit; fill the oil cup of the apparatus, to a point one-quarter of an inch below the top of the same, with the oil to be tested; when the said oil is at a temperature under 80 degrees Fahrenheit, suspend a round bulb thermometer, graduated in single degrees Fahrenheit, so that the bulb is just submerged in the oil to be tested and is in the center of the surface of the oil; heat the water-bath with a lamp, the flame of which can be regulated, so that the temperature of the oil to be tested shall rise at the rate of not less than 2 nor more than 3 degrees Fahrenheit a minute until the temperature of 90 degrees Fahrenheit is reached. At this point remove the lamp, and when the temperature of the oil to be tested, as indicated by the thermometer, has reached 95 degrees Fahrenheit, try for a flash with a small bead of fire passed slowly over the surface of the oil in the cup and within a quarter of an inch from said surface. If the oil does not take fire and continue to burn, replace the lamp under the water-bath, thereby heating the oil at the previously described rate until the temperature of 95 degrees is reached; then again remove the lamp, and, when the oil has risen to the temperature of 102 degrees Fahrenheit, try for flash and burn in the same manner as at the temperature of 95 degrees Fahrenheit. Repeat these operations, trying for the burn at seven-degree intervals, until the oil in the cup takes fire and burns continuously. The temperature at which such continuous burning takes place shall be considered the 'Fire Test,' and the temperature at which the vapors from the oil first ignite, but do not burn continuously, shall be considered the 'Flash Test.'

"Section 2. That any person or persons, firm, corporation or corporations, or officer or officers thereof, violating the provisions of Section 1 of this Act, shall, upon conviction, be subject to a fine not to exceed five hundred dollars ($500.00), or imprisonment not more than six months, at the discretion of the court.

"Section 3. This act shall be in force from and after ninety days from its passage; and all acts or parts of acts inconsistent with the provisions and purposes of this act to be and the same are hereby repealed."

USE OF FUELS IN THE UNITED STATES NAVY

By H. I. Cone

U. S. N., Washington, D. C.

The fuels used in the Navy are coal, fuel oil, and gasoline.

Coal must have certain characteristics to make it suitable for Naval use.

One of the most important strategical requirements of a warship is her ability to steam great distances without recoaling. Another is her ability, in time of need, to make high speeds. As both bunker capacity and boiler power are limited by other features of design, it follows that the coal must have the greatest thermal efficiency obtainable in connection with the other necessary characteristics.

Considerations of weight and space impose certain restrictions upon boiler design. In naval boilers the ratio between cubic foot of combustion space and pounds of fuel consumed per minute is necessarily much less than in other types. The effect may be most readily grasped if expressed in terms of the length of time that a given volume of the gases generated by the burning coal remains in the furnace. The following figures are quoted from a report of the Bureau of Mines of the Interior Department:

	Fuel per hr. per. sq. ft. of grate	Time each cu. ft. of gas stays in furnace
Torpedo Boat Boiler	40 lbs.	.077 seconds
Modern Locomotive	60 lbs.	.17 seconds
Stationary (Heine)	25 lbs.	.58 seconds

It is apparent from the foregoing that naval coal must be as low as possible in volatile content. This second characteristic is as important as the first, for it matters little if the coal is high in B.T.U.S. but is of such chemical composition that too many of them group the smoke pipe in the form of unconsumed gases and tarry vapors. But it is usually difficult to get a coal producer to admit the inferiority of his product for naval use when a chemist has certified that laboratory analysis shows it to contain as many thermal units as coal that the Navy is buying, but to run perhaps 20% less in fixed carbon.

High volatile coals are usually very smoky in naval boilers. Smoke not only reveals the location of the fleet but might fatally interfere with accuracy of gunfire.

Amount of ash is a characteristic affecting not only the ultimate thermal efficiency to a greater degree than laboratory analyses may indicate, but also the amount of labor involved in disposing of the refuse. Exhaustion of the firemen may be the determining factor in battle.

Amount and nature of clinkers has the same bearing as ash. Some coals suitable in other respects fuse to the grates to an extent which soon shuts off the air supply; their removal is the most exhausting work a fireman has to do.

Coal should coke readily to avoid loss through the grates as fires have to be "worked" vigorously when steaming at high powers, and during the frequent, sudden and great changes in speed incident to fleet manœuvres.

The final characteristic is cost. It is fortunate for the Navy that the coals which best fulfill all the other characteristics are usually the cheapest in dollars and cents per horse power developed.

Extended experience under service conditions and numerous special tests show that the semi-bituminous coal from the great fields in Southeastern West Virginia, Western Maryland and Southern Pennsylvania are the American coals which best fulfill these requirements. They comprise the great bulk of all the coal used in our Navy, both at home and on foreign stations. In special cases where it is desirable or necessary for ships remote from Government colliers or Government coal piles to purchase coal, Admiralty Welsh is usually selected if American coal of the required kind is not obtainable at lesser cost. Welsh coal is not different from ours in its performance in naval boilers, but is much more likely to cause trouble from explosive gases in the bunkers.

Coal equal or superior to the American coals now used is thought to exist in great quantities in Alaska, but it seems questionable whether the fields will be developed sufficiently to supply even a part of the Navy's needs before the time when most of the coal burning ships will have been written off the Navy's books.

The advantages of oil as compared with coal are:

An evaporation per pound of fuel in the ratio of about 14 to 9, and per square foot of heating surface in about the ration of 10 to 8.

Fuel can be taken aboard more rapidly without manual labor, and without interruption to the routine of the ship. The problem of fueling at sea is solved.

Steam for full power can be maintained as readily as for low power. A vessel burning oil is capable of runs at full speed limited in duration only by the supply of fuel. There is no reduction in speed due to dirty fires or to difficulty in trimming coal from remote bunkers, or to exhaustion of the fireroom force.

There are no cinders and the amount of smoke can be controlled.

A considerable reduction in *personnel* is possible.

The weight and space required for boilers is reduced. First, by the reduction in heating surface required, and second, by the shortening of firerooms. Consequent on the reduction in heating surface is a decrease in weight and cost of boilers.

Coal and ash handling gear is eliminated. This renders unnecessary the piercing of the hull for coal trunks and discharges from the ash expellers or ash ejectors.

The stowage and handling of oil is much easier than of coal and will result in a much cleaner ship with consequent increase in time available for drills.

The mechanical supply of fuel to the boilers gives a prompt and delicate control of the steam supply, permitting more sudden changes in speed than with coal, which is a tactical advantage.

The nature of fuel oil permits utilization of remote portions of the ship and of constricted spaces for its stowage.

These advantages have long been recognized by the Navy and there have been experiments with liquid fuel dating back as far as 1867. All these experiments have confirmed our belief in the considerable military advantages which will accrue from its use, but until recently it has been impracticable to use it extensively on account of the uncertainty as to the adequacy of its supply and the sufficiency of its distribution among the seaports of the world.

We are now assured, however, as regards the supply, that there is sufficient oil on the public lands of the State of California alone to supply all probable naval demands for one hundred years should oil be burned to the exclusion of coal, and of course there is considerable oil in other portions of U. S. territory. The question as to the distribution of oil among the ports from which fuel might be required by our vessels in time of war is one that is well within our power to solve, as from its nature the oil can be transported and stored more easily than can coal. Indeed for the transport of oil in time of war we are already better provided than for coal, there being a large number of tank steamers flying the American flag.

Oil is therefore certain rapidly to replace coal as a fuel for Naval purposes.

Since 1907 all torpedo boat destroyers contracted for, of which there are 29, burn oil exclusively, and the battleships Delaware, North Dakota, Florida, Utah, Wyoming, Arkansas, Texas and New York, contracted for during this period, are fitted to burn oil as auxiliary to coal, each of these vessels carrying about 400 tons of the liquid fuel, to be burned at full power after the coal fires become dirty, or when it becomes difficult to trim coal from the bunkers into the firerooms. In the case of these battleships the advantages of the oil have so appealed to the *personnel* that oil alone is burned to a great extent in port, and to some extent while cruising, although the installation of the oil-burning equipment did not contemplate these uses.

The Nevada and Oklahoma, the two battleships which have recently been contracted for, will burn oil exclusively. This is perhaps the most radical development in Naval engineering since the advent of the turbine. It has permitted in the case of these vessels a reduction in boiler weights, which has made possible the use of heavier armor than has hitherto been employed. The reduction in length of boiler compartments has permitted the grouping of all boilers under one smoke pipe, which course clears the upper deck considerably and permits more extensive arcs of fire for the turrets.

Aside from the use of oil as fuel under steam boilers, it now seems probable that within comparatively few years oil used in internal combustion engines will furnish the principal fuel for all Naval vessels. This is in consequence of the recent remarkable development of heavy oil engines of the Diesel type in Europe. Hitherto, oil engines have not merited much consideration for large naval vessels on account of the limited power that could be developed in a single cylinder. An installation of any considerable power required a multiplicity of cylinders. Now, however, we are credibly informed that 1,000 horsepower has been developed in a cylinder about 33″ in diameter with a 40″ stroke, at 150 R.P.M. in a 2 cycle marine type readily reversible engine. This engine has a speed control that is satisfactory, and an economy of fuel consumption probably twice that of a steam engine.

In the U. S. Navy heavy oil engines built or so far projected are limited to a number of submarine vessels and to mother ships for submarines. The former develop 1200 horse power, distributed between two shafts; the latter 900 horse power on one shaft.

Gasoline is used as fuel for all of our earlier submarines and for a large number of small power boats carried by warships. Its use is likely to be discontinued entirely as soon as suitable heavy oil motors for the small power boats are developed. As stated above heavy oil engines are already supplanting gasoline engines in submarines.

THE OXIDATION AND DETERIORATION OF COAL

By Alvin J. Cox

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Owing to the necessity of storing coal and to the fact that it is being stored in increasing amounts from year to year, it appears desirable to investigate the changes which take place when coal is exposed to the air. Probably the most notable of these changes is the direct addition of oxygen, and it is this, with its accompanying reactions, which I desire to discuss. When one considers the comparative uniformity of the composition of coals, it is not surprising that the capacity to absorb oxygen should be a common property. On the other hand, there is a decided difference between the kinds of coal, and the rate and amount of the oxygen absorption partly depends on this factor.

The literature is full of extravagant statements concerning the results of the exposure of coal to atmospheric conditions, yet much reliable data has been published on this subject and the idea is generally accepted that a change occurs in bituminous and lower grades of coal which is of sufficient magnitude to make it of industrial significance. But the economic importance of the ever-present, slow oxidation of coal is probably under-estimated, and much additional investigation must be carried on before we shall arrive at a complete understanding of all the factors which influence the deterioration of coal. It seems undesirable in this paper to discuss the literature of the subject in detail, and I shall confine myself chiefly to the results of my own experimental work. I began this study in 1908. I have selected samples of coal for my investigation from some of the best sources in the Philippines.

Sample No. 1 is a coal from the Military Reservation, Batan Island, several tons of which were shipped in sacks direct from the mine to the Bureau of Science for a steaming test. During the test, one hundred kilograms were collected, a shovelful at a time, and from this a small sample was secured by quartering and reducing the size of the pieces to that of a small marble. A two-liter glass-stoppered bottle was completely filled from the sample and the air excluded. This was reserved for experimental purposes.

Sample No. 2 is from Polillo and was taken by me from a pile of 16 tons which had lain in an open bunker for a year. The sample was prepared and kept as described for No. 1.

Sample No. 3 is from Comansi Vein No. 2, near Danao, Cebú. The coal was shipped in sacks from Cebú, soon after being mined, direct to the Bureau of Science for a steaming test. The sample was taken and bottled as described for No. 1.

Sample No. 4 was brought to the laboratory from Bulalacao, Mindoro, in a sack. This coal was sampled as above described.

Sample No. 5 came from the mountain district of Luzon and had been stored in sacks in our warehouse for one and one-half years, when it was sampled for these experiments in the same manner as the others.

At the beginning of the experiments, each sample was carefully worked over to remove non-coal particles, cracked until all pieces would pass a sieve having two meshes to the centimeter (five meshes to the inch), and that portion which would pass a sieve having four meshes to the centimeter (ten meshes to the inch) was discarded. It was then looked over again grain by grain to see that no earthy matter or lint was intermixed. The samples were made as uniform as possible. All these operations were carried on in the air. For each experiment a sample was quartered from the bulk sample, so that all results are comparable in this respect. All analyses were made according to standard chemical methods and especial care was exercised to maintain uniform conditions. In the determinations of the calorific value, the same Berthelot-Mahler bomb calorimeter was used throughout and the temperature differences were determined by the use of a Beckmann thermometer. I have made a water correction of the results by subtracting six calories for each per cent of water. All corrections are comparable, and I believe that irregularities in the samples and manipulation could not have produced errors of any consequence.

Two 250-gram samples of each coal, one in air and the other in oxygen,[1] were stored for about nine months in seven-liter bottles at about 30° C. in an ordinary room lighted by diffused sunlight. The bottles were stoppered with rubber stoppers through each of which passed two glass tubes by means of which the gas samples were taken. Fresh air was frequently supplied with a bulb to those bottles containing air. The small size of the coal particles offers a greater surface in proportion to the mass and thus facilitates the absorption of oxygen. Analyses of the samples before and after storage are as follows:

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[1] Ordinary commercial oxygen, free from carbon dioxide, 91.5 per cent of which was absorbed in alkaline pyrogallol was used. The bottles were alternately partially evacuated and filled with oxygen from the bomb, until the gas left in the bottles was practically commercial oxygen.

TABLE I.—ANALYSES AND CALORIFIC VALUES OF PHILIPPINE COALS STORED IN AIR AND OXYGEN RESPECTIVELY

COAL NUMBER	ORIGINAL SAMPLE					AFTER STORAGE IN AIR			AFTER STORAGE IN OXYGEN		
	Moisture	Volatile combustible matter[1]	Fixed carbon	Ash[2]	Calories[3]	Moisture	Ash	Calories	Moisture	Ash	Calories
	Per cent	Per cent	Per cent	Per cent	Per cent	Per cent	Per cent	Per cent	Per cent	Per cent	
1	5.90	46.30	40.08	7.72	6159	5.59	7.52	6119	5.75	8.11	6059
2	8.61	40.67	46.55	4.17	6012	8.75	4.50	5931	8.03	4.25	6022
3	11.55	37.43	46.95	4.07	6066	11.11	4.47	5985	11.18	4.20	5982

The above calorific values are not comparable as they stand, since the samples contain different quantities of diluents (water + ash[4]).

When they are calculated to an ash- and moisture-free basis (coal—(water + ash)), we then deal with alterations in the pure coal substance, and the results show that in each case the calorific value is lowered by storage as given in the following table:

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[1] After the method of Cox, *Phil. Journ. Sci.*, *Sec. A* (1907), *2*, 41.

[2] Each analysis shows somewhat less ash than the ordinary coal from the same source because of the way in which the samples were prepared.

[3] Average of three closely agreeing, independent determinations.

[4] This does not represent the original amount of ash with its hydrated silicates and with its iron pyrites instead of ferric oxide formed by burning (Parr & Wheeler, *Journ. Ind. & Eng. Chem.* (1909), *1*, 636), but the same coal burned to ash in the same way unquestionably gives concordant results.

TABLE II.—THE DETERIORATION IN CALORIFIC VALUE OF COALS STORED IN AIR AND IN OXYGEN

Coal Number	Calorific Value of the Pure Coal			Per Cent Lowering of Calorific Value	
	Original sample	After storage in Air	After storage in Oxygen	Stored in Air	Stored in Oxygen
1	7130	7042	7034	1.2	1.3
2	6893	6836	6865	0.8	0.4
3	7189	7089	7069	1.4	1.7

The gases above the coal at the end of the storage period were analysed as follows:

TABLE III.—COMPOSITION OF THE GASES ABOVE THE COALS STORED IN AIR[1]

Coal Number	Volume Percentages		
	Carbon Dioxide	Oxygen	Remainder
1	1.5	18.7	79.8
2	0.2	17.7	82.1
3	1.3	15.8	82.9
4	2.2	15.9	81.9
5	3.2	15.7	81.1

[1] The gases above the coal were frequently replaced with air freed from carbon dioxide to insure an adequate supply of oxygen. Therefore, these results do not show the total amount of carbon dioxide evolved but show its presence and the exact composition of the gas above each coal when it was removed for analysis.

TABLE IV.—COMPOSITION OF THE GASES ABOVE THE COALS STORED FOR NINE MONTHS IN COMMERCIAL OXYGEN

Coal Number	Volume Percentages		
	Carbon Dioxide	Oxygen	Remainder[1]
1	6.0	22.9	71.1
2	1.6	19.5	78.9
3	2.6	14.2	83.2
4	4.5	17.9	77.6
5	4.9	14.4	80.7

The glass tubes carried by the rubber stoppers of the storage bottles were closed with rubber tips which, as well as the stoppers themselves, deteriorate so rapidly in a tropical climate that it was impossible to keep the bottles tight. The analyses indicate that as fast as oxygen was absorbed by the coal it was replaced by air. The bottles which contained the coal stored in oxygen were refilled with oxygen after they had been sampled for the analyses of Table I and after fifteen months the composition of the gases above the coal samples were as follows:

TABLE V.—COMPOSITION OF GASES ABOVE COAL IN SECOND STAGE OF STORAGE IN OXYGEN

Coal Number	Volume Percentages		
	Carbon Dioxide	Oxygen	Remainder
1	14.6	42.9	42.5
2	10.3	39.8	49.9
3	25.7	29.1	45.2
4	14.4	30.7	54.9
5	63.0	7.2	29.8

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[1] Unabsorbed in alkaline pyrogallol. Traces of carbon monoxide were detected, but hydrogen, methane, formaldehyde, formic acid, or hydrogen peroxide were not recognized by ordinary means of identification as reaction products, although it is probable that some or all of them were present in small quantities.

These results show further deterioration of the coal samples and, taken with these of the preceding table, indicate that the evolution of carbon dioxide is not simultaneous with the absorption of oxygen but is subsequent to it.[1] It is possible that a number of oxidation processes varying in character and speed of reaction are involved, depending on the kind of coal, the stage of the process, etc., but the facts are satisfactorily explained by the assumption that oxygen is absorbed by the unsaturated chemical compounds of the coal to form organic peroxides, and carbon dioxide is evolved when the peroxides break down.[2] In cases of autoxidation, hydrogen peroxide has been frequently met with and it is probable that in the autoxidation of coal the organic peroxides may be hydrolyzed slowly by water to form hydrogen peroxide which reacts upon the coal to produce carbon dioxide.

There is a marked increase in the weight of Philippine coal when exposed to the air, although no definite quantitative determinations have been attempted on account of the fluctuation of the moisture content. Absorbed gases could reduce the calorific value of coal by simply adding weight to it, *i.e.*, they would be merely a diluent. However, if oxygen on being absorbed enters into the composition of the coal or unites with the carbon or hydrogen of the coal, there is actual deterioration. When pure coal becomes oxidized, the total weight and the total heating value are decreased without changing the calorific value of what remains.

The avidity with which coals absorb oxygen is shown by the following experiments. Samples of one hundred grams each were sealed in glass bulbs in an atmosphere of air or oxygen as indicated. After a period of twelve months, the pressures were measured and the gases analyzed as follows:

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[1] Porter, H. C., and Ovitz, F. K., *Journ. Am. Chem. Soc.* (1908), *30*, 1486, note, "Some coals rapidly absorb oxygen from the air surrounding the coal during storage without forming carbon dioxide;" however, the maximum duration of their tests was 104 days and from their own numbers the content of carbon dioxide began slightly to increase after thirty-four days. Also *Ibid.* (1910), *2*, 80, "Coal absorbs oxygen from the air during storage without forming carbon dioxide and the amount of oxygen absorbed accords approximately with the deterioration in heat value."

[2] Brooks, B. T., *Phil. Journ. Sci., Sec. A* (1910), *5*, 219, offered this hypothesis to explain the oxidation of Manila copal by the air.

TABLE VI.—ANALYSES OF RESIDUAL GASES, WHEN COAL IS STORED WITH RELATIVELY SMALL AMOUNTS OF OXYGEN

COAL NUMBER	STORED IN	DIMINISHED PRESSURE IN MM.	VOLUME PERCENTAGES OF THE GASES AFTER STORAGE		
			Carbon Dioxide	Oxygen	Remainder
1	air	123	5	0.4	94.6
2	oxygen	504	75	0.0	[2]25.
3[1]	air	106	7	0.0	93.
3	oxygen	600	59	0.0	[2]41.
4	air	70	13	0.0	87.
5	oxygen	167	92	0.0	[2] 8.
x[3]	air	120	8	0.0	92.

The above numbers show that the absorption of oxygen by coal is complete when an insufficient supply of the former is present.

An apparatus to measure the rate of absorption of oxygen by coal, which consists of a barometer for measuring the pressure within a hermetically sealed space, was devised and constructed as shown in Figure 1.

The portion *a b d e* was made of capillary tubing and then at *a* sealed onto an ordinary 100-cubic-centimeter glass pipette *f*. The tube *b c* is about 800 millimeters long. The apparatus was inverted, the coal sample inserted through the wide tube *g* which was afterwards sealed in a gas flame and allowed to cool, and finally the apparatus was returned to its upright position. The heavy rubber tubing, *d h*, is of sufficient length to allow free movement of the mercury vessel *i* over the distance *b c*. The vessel *i* was partially filled with mercury and then lowered below *c*. The apparatus was alternately partially evacuated and filled with commercial oxygen (6.4 cubic centimeters not absorbed in alkaline pyrogallol) at *e*, until the gas left in *f* had practically the above composition. The vessel *i* was next raised until the mercury in it

[1] Vein No. 3.

[2] This is accounted for by the inert gas in the commercial oxygen.

[3] From the East Batan Coal Company Mine.

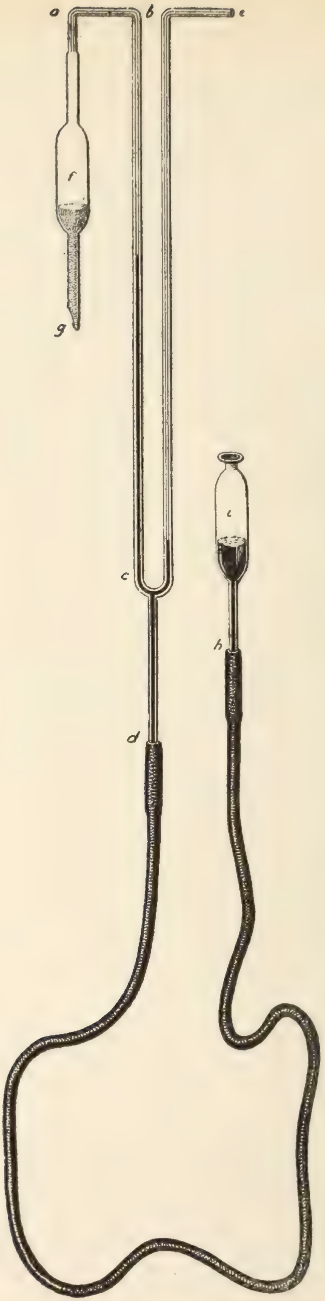

Figure 1

An Apparatus to Measure the Rate of Absorption of Oxygen by Coal was level with that in the tube *c e*, and the difference in height of the mercury in *c b* and *c e* noted. Simultaneous barometric and temperature readings were always made and from these the exact gas pressure in *f* was calculated.

The samples used in all of the foregoing experiments had been exposed to the air for some time previous to the beginning of the experiment. The deterioration, which unquestionably begins with the mining of the coal, consists in the loss of combustible gases, and oxidation already must have progressed considerably, and therefore the amount of oxidation found by the analyses is not nearly as great as it would have been if the measurements had been begun with fresh samples. Therefore, a sample from a new face of the coal from the mine of the East Batan Coal Company was secured for the following absorption experiments and brought to the laboratory in a sealed container. A portion of the sample was ground to pass a sieve having 2 meshes to the centimeter (5 meshes to the inch) and graded into the following sizes:

Portion	Passed Sieve		Held on Sieve		Weight of fraction in grams
	Meshes per cm.	Meshes per inch	Meshes per cm.	Meshes per inch	
A	2	5	7	10	2005
B	4	10	8	20	1080
C	8	20	24	60	735
D	24	60	40	100	50
E	40	100	60	150	50
F	60	150	. . .	. . .	75

Analyses of these portions are as follows:

TABLE VII.—ANALYSES OF PORTIONS OF A PHILIPPINE COAL PASSING SIEVES OF DIFFERENT SIZES

(Numbers give Percentages)

	A	*B*	*C*	*D*	*E*	*F*
Moisture	22.82	22.37	23.04	22.51	21.85	21.96
Volatile combustible matter	33.64	34.32	33.28	33.32	33.19	33.67
Fixed carbon	39.04	38.33	38.66	38.34	39.15	37.77
Ash	4.50	4.98	5.02	5.83	5.81	6.60
Total	100.00	100.00	100.00	100.00	100.00	100.00
Color of ash	Dark brown	Yellowish brown	Yellowish brown	Yellowish brown	Dark brown	Dark brown

The relative rate of the absorption of oxygen was obtained for a 20-gram sample of each of the following, namely, *A* quickly ground in a mortar which I will designate *AA*, *A*, *C*, *E*, and *F* when each was placed in a piece of apparatus like Figure 1. About two hours were required to crush, sift, and weigh the portions and samples, except *AA*, and to exhaust, seal, and begin the actual measurements. The results are as follows:

TABLE VIII.—THE COMPARATIVE AVIDITY OF COAL PARTICLES OF DIFFERENT SIZES FOR OXYGEN AT 30° C.

TIME AFTER STARTING	DIMINISHED PRESSURE IN MILLIMETERS				
	AA	*A*	*C*	*E*	*F*
18 hours	270	109	270	253	165
26 hours	326	147	333	273	224
42 hours	399	191	407	355	295
3 days	460	251	472	452	363
4 days	495	313	505	493	406
5 days	514	343	520	505	432
7 days	**521**	400	**551**	**516**	**456**
10 days	519	452	523	507	454
36 days	503	**544**	501	485	447
365 days	477	518	456	462	386

It is easily seen from the above table that with each coal the pressure diminishes for a time and then increases. The reduction in the pressure was due to the gradual absorption of oxygen by the coal, but in time the rate of the evolution of carbon dioxide was equal to that of the absorption of oxygen, and later even exceeded it and thereby increased the pressure. These observations substantiate the assumptions made in considering Tables IV and V, that the evolution of carbon dioxide is subsequent to the absorption of oxygen.

The absorption of oxygen by *A*, the coarsest sample, after sealing is not very rapid; but that by *C*, which is very much finer than *A*, is 2½ times as fast. Sample *AA* is the same portion as *A*, but was quickly pulverized in a mortar immediately before beginning the experiment, and in this sample the absorption in the sealed apparatus was 2½-fold greater than *A*. On account of the great avidity for oxygen shown by the finely divided coal, it was anticipated that samples *E* and *F* which consist of the finest particles, on account of the small amounts of these portions obtained in the sizing and consequently excessive exposure to the air during preparation, might be fairly saturated before the experiment began. This was the case as indicated by the low speed of absorption of oxygen after sealing and the comparatively small reduction in the pressure before it began to increase. The fact that not so great a partial vacuum was produced at any time in these as in the fresher samples shows that the rate of the evolution of carbon dioxide was equal to that of the absorption of oxygen while the latter was still quite energetic. The evolution of carbon dioxide depends directly on the previous absorption of oxygen and therefore the fact that the evolution of carbon dioxide by samples *E* and *F* was more rapid than that in any of the others clearly substantiates the assumption as to their previous partial saturation.

The rate of the absorption of oxygen by a given coal varies with its previous exposure to the air. Under the same conditions, it depends upon the size of the grains; it is more rapid in the finer particles, probably approximately proportional to the surface exposed. The absorption of oxygen, not including that replaced by evolved carbon dioxide, was determined in two samples each of portions *A* and *C* in apparatus identical with that shown in Figure 1 and described above except that flasks of dimensions as indicated were substituted for the 100-cubic-centimeter glass pipette *f*.

TABLE IX.—THE OXYGEN, NOT INCLUDING THAT REPLACED BY CARBON DIOXIDE AND OTHER EVOLVED GASES, ABSORBED BY BATAN COAL AT 30° C.

Time in days	Portion *A* (40 Grams in 4.390-Liter Flask [1]			Portion *A* (100 Grams in 4.245-Liter Flask [1])			Relative Rates of Absorption of the 2 Samples of Portion *A*	Portion *C* (20 Grams in 1.394-Liter Flask [1])			Portion *C* (40 Grams in 1.546-Liter Flask [1])			Relative Rates of Absorption of the 2 Samples of Portion *C*.
	Volume[3] of Oxygen Absorbed in Cubic Centimeters per Gram Coal	Per cent by Weight of Oxygen Absorbed Referred to the Fresh Coal	Pressure in Flask in mm.	Volume[2] of Oxygen Absorbed in Cubic Centimeters per Gram	Per cent by Weight of Oxygen Absorbed Referred to the Fresh Coal	Pressure in Flask in mm.		Volume[2] of Oxygen Absorbed in Cubic Centimeters per Gram	Per cent by Weight of Oxygen Absorbed Referred to the Fresh Coal	Pressure in Flask in mm.	Volume[2] of Oxygen Absorbed in Cubic Centimeters per Gram Coal	Per cent by Weight of Oxygen Absorbed Referred to the Fresh Coal	Pressure in Flask in mm.	
0	0.00	0.00	754	0.00	0.00	783	......	0.00	0.00	7.67	0.00	0.00	770	......
1	1.00	0.13	747	0.99	0.13	765	1:0.99	1.81	0.23	7.47	1.80	0.23	734	1:0.99
5	2.15	0.28	739	2.14	0.28	744	1:0.99	4.44	0.57	7.18	4.34	0.56	683	1:0.98
13	4.30	0.55	724	4.17	0.54	707	1:0.97	7.26	0.94	6.87	7.09	0.91	628	1:0.98
21	6.17	0.80	711	5.92	0·76	675	1:0.96	9.51	1.23	6.61	9.33	1.20	583	1:0.98
31	7.75	1.00	700	7.40	0.95	648	1:0.95	11.07	1.42	6.45	0.63	1.37	557	1:0.96
46	10.05	1.29	684	9.38	1.21	612	1:0.93	13.51	1.74	6.18	2.77	1.64	514	1:0.95
59	11.77	1.52	672	10.70	1.38	586	1:0.91	14.69	1.89	6.05	3.82	1.78	493	1:0.94
65	12.77	1.65	665	11.29	1.45	577	1:0.88	15.15	1.95	5.99	.....	.....	...	....

[1] Gas volume in flask in c.c.—volume of flask in c.c.—(weight coal in grams × specific gravity coal). The specific gravity used in the calculations 1.3.

[2] Computed to standard conditions of pressure.

[3] Experiment in progress.

In the larger samples the absorption was so much more rapid that the pressure at which the oxygen was supplied became diminished and the rate of absorption correspondingly reduced. The particles in the samples of the portions were so fine that they packed closely, and particularly in the larger samples the oxygen possibly could not penetrate uniformly to all parts. The relative rate of the absorption of oxygen in the two samples of portion *A* and of *C* was never equal to the ratio of their weights. The above experiment also shows that the absorption of oxygen is much more rapid during the first few weeks than during the subsequent periods. These points are indicated more clearly when the results of Table IX are represented graphically as shown in Figure 2 in which Curve I = Portion *A* (40 grams in 4.390-liter flask); Curve II = Portion *A* (100 grams in 4.245-liter flask); Curve III = Portion *C* (20 grams in 1.394-liter flask); and Curve IV = Portion *C* (40 grams in 1.546-liter flask).

If coal is stored in air, after a time the compounds in the coal unsaturated with respect to oxygen become saturated and the avidity of the whole for oxygen becomes much less as shown in the two preceding tables. It follows from this that there is much more danger from fire during the early stages of storage, and because of this fact, when coal has once been stored as a reserve and no serious heating has resulted, it should be left undisturbed and the coal for present consumption taken from the newest piles.

The value of the bituminous coal produced in the United States during the calendar years 1907 to 1909 was $1,230,836,887, an average annual value of $410,278,962.[1] If bituminous coal suffers an average deterioration of two per cent[2] the annual loss in the United States from deterioration of coal is over eight million dollars, or four per cent as great as the total losses of all property by fire.[3]

[1] U. S. Geol. Surv. *Min. Pred. U. S. Calendar Yrs. 1900–9* (1911), Mar. The average annual production of Pennsylvania anthracite is valued at $157,059,584.

[2] Parr, S. W., and Hamilton, N. D., *Econom. Geol.* (1907), *2*, 703, found that in 20-pound lots of Illinois coal "outdoor exposure results in a loss of heating value varying from 2 to 10 per cent" which substantiates Table IX. *Also cf. Engineer* (London) (1903), Oct. 30, and Bement, A., *Chem. Eng.* (1910), *12*, 10.

[3] From the data of the National Board of Fire Underwriters, U. S. Dept. Commerce and Labor, Bur. Statistics, *Statis. Abs. U. S.* (1910), 586, the estimated fire losses in the United States for the calendar years 1907 to 1909 aggregate $621,675,709, an average annual value of $207,225,236.

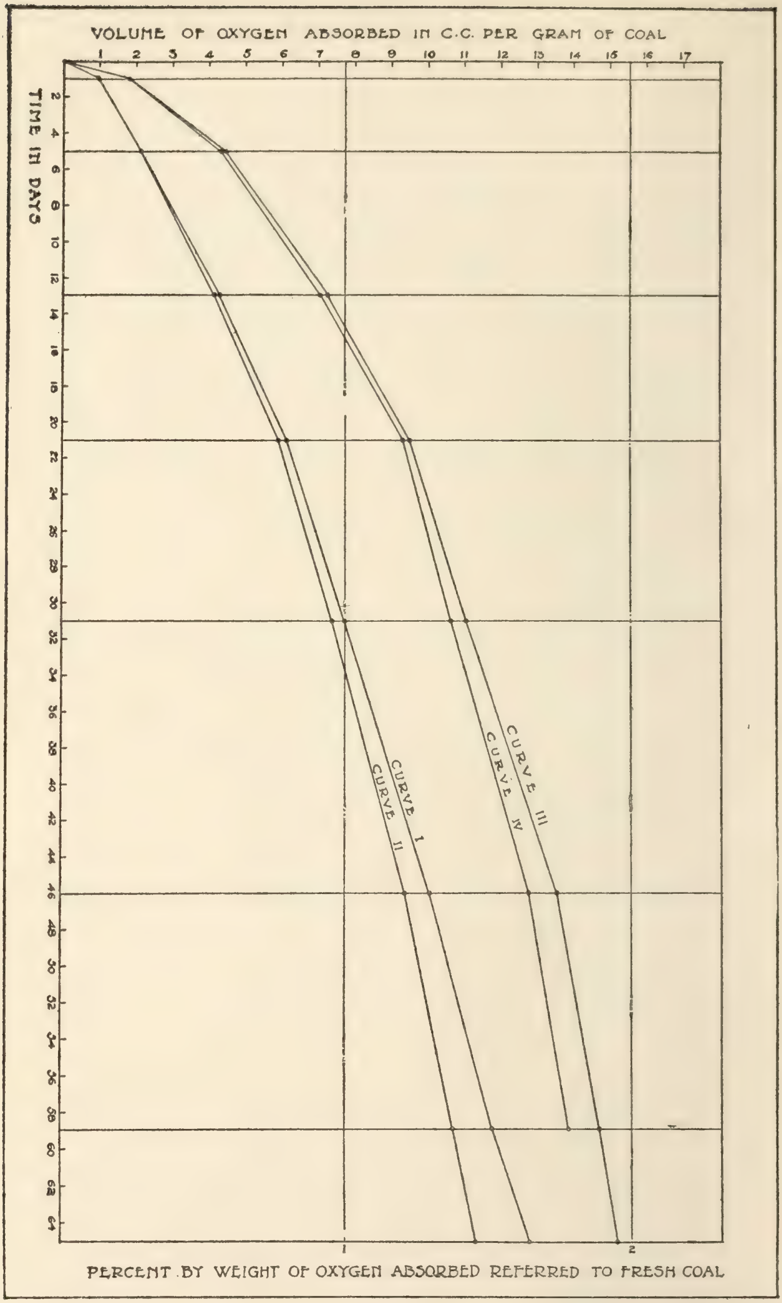

Figure 2.

Curve Representing the Oxygen, Not Including that Replaced by Carbon Dioxide and Other Evolved Gases, Absorbed by Batan Coal at 30° C.

There is also a large loss due to spontaneous combustion of coal. Oxidation of combustible matter produces the same amount of heat whether the reaction takes place rapidly or very slowly. If the heat of a slow reaction can not escape, the same increase in temperature will result as when the change takes place rapidly. Every combustible substance has its ignition point, and for it to burn rapidly it first must be heated to the point of ignition. This depends almost entirely on the porosity and the fineness of division, together with the chemical composition. If the substance is in a large compact unit, its ignition is not easily accomplished. But in a very finely divided state and with sufficient oxygen present, it may kindle and the combustion be propagated with explosive violence. If the amount of coal in a given heat be small, there will be no perceptible increase in the temperature from the heat of the reactions involved, for the heat will all be lost through radiation. But if the escape of heat is prevented, as is done when fine coal is piled in high heaps, and if a supply of oxygen is available, absorption of oxygen with consequent rise in temperature will result. The absorption of oxygen by coal seems to increase rapidly with the temperature, and eventually the kindling point may be reached and visible combustion follow.[1] In an extremely finely divided state coal will ignite almost as readily as gas derived from coal. Care must therefore be taken to avoid the ignition of coal dust, as there is no doubt that many serious fires and explosions are directly attributable to it, and the violence of the latter may have been more severe in certain cases on account of absorbed oxygen.

The presence of sulphur in the form of iron pyrites in stored coal has often been thought to be a source of heat and the cause of spontaneous combustion owing to the reaction between sulphur and oxygen.[2] It is a fact that coals with a high content of sulphur often give trouble from heating; on the other hand, coals containing a small amount of sulphur, not more than 1½ per cent, which include practically all of those from the Philippine Islands, also sometimes heat badly. This can not entirely be explained by the sulphur present. The amount of sulphur is far too small to account for the increase in temperature recorded in many instances even if it were all oxidized, which has never been shown to be the case. Porter, H. C., and Ovitz, F. K.,[1] have shown that there is "a very minor contribution, if any, on the part of sulphur to spontaneous heating in coal." Bodenstein, M., and Karo, W.,[2] have shown that for liquid sulphur the velocity of the reaction with oxygen is proportional to the surface exposed. On the same principle, the fineness of division of the sulphide in coal would make its oxidation comparatively rapid and favor spontaneous combustion.

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[1] Ordinarily the air furnishes the oxygen for combustion, but it may be supplied by other oxidizing agents. In the latter case oxidation may be so rapid that it causes an explosion. Certain Philippine coals mixed with sodium peroxide at room temperature ignite spontaneously.

[2] Cf. Parr, S. W., and Kressman, F. W., *Jour. Ind. & Eng. Chem.* (1911) *3*, 151.

The question of the influence of moisture in coal[3] upon its oxidation has not been sufficiently worked upon to enable us to decide it. Griswold says that when stored, wet coal should be spread out to expedite drying, especially when wet with snow and ice, and Doan[4] says, "The amount of moisture in an air-dried sample of coal is an indication of its liability to heat, the danger increasing with the percentage of moisture contained. Coal containing over 4.75% after air drying is said to be dangerous." The amount of water naturally present in coal is an indication of the grade and, as already pointed out, the kind of coal is a factor upon which oxidation partly depends. Aside from this, it is probable that the amount of moisture has little significance.

The action of sunlight on coal not only increases its temperature directly, but also catalytically by accelerating the absorption of oxygen, and thus in two ways it increases deterioration and the danger from spontaneous combustion.

A coal which illustrates these points is that now being mined on the eastern end of the Island of Batan.[5] On exposure to the sun and air it alters rapidly, loses its luster, and falls into powder. It slakes in shipping, unless it is kept from the sun and wind while in transit. Spontaneous combustion has several times occurred in this coal, but the particulars are not available, except in the more

[1] *Journ. Ind. & Eng. Chem.* (1912), *4*, 7.
[2] *Ztschr. f. phys. Chem.* (1910), *75*, 47.
[3] *Eng. News* (1904), *51*, May 5.
[4] *Eng. News* (1904), *52*, 141.
[5] This coal is a poorer grade than that from the western end of the Island.

recent cases. The Civil Government used some coal from the Batan mine during 1911 and had trouble with 9000 tons of the same coal in piles about five meters high placed on the ground under a roof. The piles were ventilated with alternating tiers of horizontal air passages, about six meters apart, running at right angles to each other. In this case there was no actual flame, but all of the timber used to make the ventilators was badly charred.

In coal heating in bunkers, even though the temperature be only moderately raised, there is an increased escape of volatile combustible matter and consequent loss of fuel value. Evidently the danger from spontaneous combustion of coal may be reduced by assisting in every possible way the dissipation of heat through radiation by having the coal sufficiently close to air-cooled surfaces,[1] but any system which brings the air freely into contact with the coal should be avoided. Ventilation of this sort may mean danger of fire, because it is very difficult to provide sufficient radiation to remove the heat as fast as it is formed. Storage piles should be protected from the sun and every other external source of heat.

Any method which protects the coal from the air will preserve it. This can unquestionably be done by submergence under water, but this necessitates firing wet coal.[2] Firing of wet coal may be avoided by dry storage in carbon dioxide or other inert gas. I believe there will be little danger from fire in storage of this sort for the air supply is cut off. Deterioration probably may be as effectually avoided by the use of bunkers, with tight floors and sides and fairly close fitting covers, filled with carbon dioxide as by storage under water. The following experiments show the effectiveness in preventing deterioration by the exclusion of air through inert gases. Two 250-gram samples of each coal, one in nitrogen and the other in carbon dioxide, were stored for about nine months in 10-liter bottles at about 30° C. in an ordinary room lighted by diffused sunlight.

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[1] The coal should be stored in bunkers which have surfaces large in proportion to the bulk, or air might be circulated through the heaps in iron pipes in such a way that it does not come in contact with the coal.

[2] Parr, S. W., and Hamilton, N. D., *Econom. Geol.* (1907), *2*, 703, say, "Submerged coal does not lose appreciably in heat value." *Also cf.* bibliography.

TABLE X.—ANALYSES AND CALORIFIC VALUES OF PHILIPPINE COALS STORED IN NITROGEN AND IN CARBON DIOXIDE

Coal Number	Original Sample					After storage in Nitrogen			After storage in carbon dioxide		
	Moisture	Volatile combustible matter[1]	Fixed Carbon[2]	Ash	Calories[3]	Moisture	Ash	Calories	Moisture	Ash	Calories
	Per cent	Per cent	Per cent	Per cent		Per cent	Per cent		Per cent	Per cent	
1	5.90	46.30	40.08	7.27	6159	5.48	7.66	6227	5.56	6.97	6249
2	8.61	40.67	46.55	4.17	6012	8.46	4.05	6050	7.68	4.91	6057
3	11.55	37.43	46.95	4.07	6066	10.39	3.81	6199	10.86	3.88	6161

The above results calculated to the uniform basis of pure coal show that there was no deterioration on storage.

TABLE XI.—THE CALORIFIC VALUE OF COALS STORED IN NITROGEN AND IN CARBON DIOXIDE

COAL NUMBER	CALORIFIC VALUE OF THE PURE COAL[4]			VARIATION FROM ORIGINAL SAMPLE IN CALORIES	
	Original Sample	After storage in nitrogen	After storage in carbon dioxide	Stored in nitrogen	Stored in carbon dioxide
1	7130	7168	7144	+38	+14
2	6893	6914	6929	+21	+36
3	7189	7225	7226	+36	+37

Table XI shows an increase in calorific value during storage. I can not explain this, but it is probable that it is due to the evolution of carbon dioxide rather than to experimental error. That such a change does take place in a coal which has absorbed oxygen subsequent to the removal of the oxygen supply is shown by the following facts.

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[1] After the method of Cox, *Phil. Journ. Sci. Sec. A* (1907), *2*, 41.

[2] Each analysis shows somewhat less ash than the ordinary coal from the same source because of the way in which the samples were prepared.

[3] Average of three closely agreeing, independent determinations.

[4] An interesting comparison is that of Table XI with Table II, from which it will be seen that the coal stored in air has a calorific value about 2 per cent less than that stored in either of the above inert gases.

An analysis of the residual gas above coal number 1 stored in nitrogen gave the following results:

	Per Cent
Carbon dioxide	0.6
Oxygen	0.0
Nitrogen	99.4

To demonstrate beyond question that the small amount of carbon dioxide did not result from a slight leakage of air through the rubber stopper and subsequent oxidation of coal, proportionately larger portions of coal numbers 1 and 2 were sealed off in an atmosphere of nitrogen in flasks. After one year they were opened and in each case the pressure was found to be increased by a small amount and the gases had the following compositions:

	From Flask Containing Coal No. 1 Per Cent	From Flask Containing Coal No. 2 Per Cent
Carbon dioxide	4.0	5.0
Oxygen	0.0	0.0
Nitrogen	96.0	95.0

The same sort of deterioration as I have just described takes place along the outcrop of coal beds and is limited only by the nature of the coal and the penetration of the air. The latter is not very great in well-watered regions like the Philippine Islands where the ground water table is high, but in arid regions it is natural to suppose that the air may penetrate and deterioration take place to proportionately greater depths. However, recent work has shown that deterioration does not always extend to the ground water level or even to a considerable portion of the distance.

This paper gives results which are by no means complete, and leaves several points unsettled and much to be accomplished. Only the preliminary investigation has been carried on, but with the information and experience derived from it the author hopes more effectively to continue the work.

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COAL-WASHING EFFICIENCY

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The term efficiency as applied to coal washing is one which had but little significance to that branch of the engineering profession. The reason of this is probably due more to the comparatively recent application of this process to coal of inferior quality than to the intentional disregard of those engaged in this work to the necessity of the universal adoption of a standard definition of coal-washing efficiency.

It was only a few years ago that only the seams of high-grade coal were mined, but the increased demand for coal caused by the wonderful growth of the manufacturing business in the last fifteen years, and the fact that practically all such coal land came under the control of the great corporations, has made it necessary and profitable to mine coal of inferior grade and, by washing, remove the impurities and thereby produce a washed coal often of equal quality to that of the best grades. This has been made possible in two ways. Late years have brought forth some wonderful improvements in the machinery used both in the mining and in the preparation of coal, resulting in greatly decreased cost of production; and this with the comparatively small acreage of high-grade coal land not controlled by the corporations, and the possibility of obtaining coal lands of an inferior grade of coal at a comparatively low figure, have made the final cost of mining and preparing such coal sufficiently low to successfully compete with the coal of better quality.

As is nearly always the case with any new industry, more attention was at first given to the development of the necessary equipment required in washing coal than to the perfection of the separations accomplished, though, of course, all engaged in the manufacture of such machinery have constantly improved their products in their competition in trade with one another. Still, it has been the custom to permit the designing of coal-washing plants to be done by these manufacturing concerns, and as they are very naturally interested in, and in fact make their profits from, the machinery with which the plants are equipped, it is only to be expected that the advancement along the lines of simplicity, convenience and high efficiency would be slow. It is not surprising then that no standard method of calculating coal washery efficiency has been adopted and this cannot be hoped for until the designing of these plants falls into the hands of engineers whose whole object is not only to produce plants that will wash the coal and remove most of the impurities, but will attain the very highest efficiency possible.

I would not wish to be understood to insinuate that the present manufacturers of coal-washing machinery do not employ competent engineers, or that their plants are in all cases inefficient, as such is not the case. Many plants recently built are very convenient, complete and efficient but, as there are no standard formulæ or methods of calculating washery efficiencies, it is impossible to intelligently compare the plants as designed by these concerns or any one else. As a result of these conditions it is practically impossible for the coal company official to intelligently determine just what type of coal-washing machine is best adapted to his particular coal. Were it possible for him to obtain data showing the efficiency of different types of machines on all different kinds and sizes of coal, and were all this data based on and calculated from a standard method and formulæ, it would then be possible for him to determine just what type of machine is best adapted to his own proposition and a great deal of expense and trouble would be averted. It would also result in a higher general efficiency in coal washing over the country, for the present haphazard method results in heavy coal losses which, I think, every engineer is becoming more and more in favor of eliminating as much as possible. As each manufacturer of coal-washing equipment has his own particular kind and style of machinery, and as no one type will handle all kinds, grades and sizes of coal with equal effectiveness, but instead will be more particularly adapted to some one kind or size of coal, it is but natural to suppose that such misfits would result when the coal company official is so much in the dark as to the efficiency of different machines on different kinds of coal that he must place his order more with a view to cost than to the probable perfection of the preparation of his coal. This first cost may often result in a much greater expense to his company before the plant will successfully wash their coal. One cannot blame the manufacturer selling the machinery in such cases for it is but natural to suppose that he would place his products wherever possible, and particularly so when he knows that practically every buyer is almost absolutely in the dark in-so-far as comparative data is concerned which would permit of his ever actually knowing that much better results could be obtained by the use of another type of machine. The question then would appear to be, how can the coal company engineers be brought to a realization that it is possible to calculate washery efficiencies and to make intelligent comparison of washery results, and that it is through their own endeavors along this line and their contributions to the engineering journals and societies that the resulting interchange of efficiency data will result beneficially to all? Is it not true that these are the very men that can force the issue and make the washery firms get their data in comprehensive form? Admitting this, is it not also a fact that some standard method of calculation of efficiencies must be adopted? To attain this, probably the best and quickest way is for the engineering societies to take up this question as they are in a better position to determine just what method of calculation should be employed, and when approved and adopted by some noted society and published in their proceedings the same would be copied by many of the engineering journals of the world and a big advancement would have been made toward the universal adoption of a standard in coal washing calculations. The writer therefore seriously hopes the reading of this paper before your society will create some interest in this subject, and result in some steps being taken along the lines suggested. There are many engineers that have but little idea of coal washing on account of never having had occasion to investigate that phase of the preparation of coal but, as nearly all are becoming more and more interested in the absolute necessity of reducing to a minimum all losses of fuel, and as it is a well-known fact to those engaged in coal-washing engineering that washery losses in many cases are high, would it not be well to investigate this subject a little and see if some action cannot be taken to assist in any way possible that may result in reducing these heavy losses?

I have made a very careful study of this subject for the past twelve years and in fact have made a specialty of that branch of the engineering business. In my own work I have adopted a method which, after very careful consideration, I feel sure is one that accomplishes the desired results and not only makes it possible to make intelligent comparisons of washery results but also gives an absolute check on all items of a test. I beg to submit my method to the consideration of your society and sincerely hope it may receive your approval, if not in entirety, then in part, and may eventually result in your adopting resolutions of a nature that will be of assistance to those working for better results and conditions in coal washing.

A great many coal companies depend almost entirely upon analyses of the raw coal, washed coal and refuse for the determination of their washery results. Analyses alone have no value whatever in the determination of efficiency. As an example, what would be considered a low ash content for washery refuse would be 30 per cent, yet such a material could not be considered a merchantable coal. There is nothing about the analysis to indicate whether each individual piece would analyze 30 per cent ash or whether it was a result of a combination of pieces some of which might analyze 65 or 70 per cent ash and others 10 or 20 per cent ash. If a perfect separation were accomplished, there should be no free pieces of coal in the refuse which, if placed in the washed coal, would raise the ash content of it above the desired percentage. As it is an absolute fact with but very few exceptions that, as the specific gravity of coal increases, so will the ash content increase (not necessarily at any fixed ratio for all coals), the simplest method of separating out the free pieces of good coal from the refuse is by placing the sample in a liquid of a density such that the good coal will float and the refuse sink.

The use of this float test is very valuable in preliminary investigations of coal-washing propositions, for by using six or eight baths of densities ranging from 1.55 to 1.70 sp. gr. it is possible to determine just what separation is necessary in washing the coal to obtain a washed product of the desired ash (or sulphur) content. The bath with a density giving this result is the one which I term the "permissible bath," and this density or specific gravity is used throughout all of the balance of my tests upon this particular coal, whether they be tests of the raw coal or of the washed coal and refuse from it. This is very important and is the basis of my method of calculating washery efficiencies. As an example of this, I do not consider good coal as that only which has an ash content of the desired percentage, but all coal contained in the raw material, whether high ash or low, which in combination one with the other results in a combined product of the desired ash content. It will be readily understood that in such combinations some pieces of coal may have a much higher ash content than that of the total float sample of the permissible bath. From this it will be understood that by making float tests of both the washed coal and refuse, using the permissible bath exclusively, any sink obtained from the washed coal should be with the refuse, and any float from the refuse should be with the washed coal. If the analysis of the washed coal in such a case is the same as that of the raw coal float from the permissible bath test, then it will be found that the percentage of washed coal is less than the percentage of float last mentioned. The float from the refuse is good coal lost and in calculating the percentage of loss I do not figure it as a percentage of the original total raw coal, as many do, but as a percentage of the possible amount of good coal contained in the raw coal. In other words, I consider it a misnomer to say that 10 per cent of the raw coal is lost in the refuse as good coal when, for instance, the raw coal was composed of 90 per cent good coal and 10 per cent refuse matter to be discarded.

Now it will be seen that, as the coal loss decreases toward zero, so will the permissible bath sink from the refuse increase toward 100 per cent one depending entirely upon the other. Therefore, by considering the efficiency of the refuse as 90 per cent where 10 per cent float was obtained on the permissible bath test, this efficiency would cover both the actual refuse efficiency and the efficiency as to coal loss. Now assuming the refuse efficiency to be 100 per cent and the coal loss, of course, zero, yet having say a 5 per cent sink resulting from the permissible bath test of the washed coal, and as a result, a lower percentage of washed coal than should be obtained as indicated by the permissible bath test of the raw coal, it could not be said that the total efficiency was 100 per cent. Instead, by taking the washed coal efficiency as 95 per cent (100–5 per cent sink) and then figuring the total efficiency as an average of both the washed coal and refuse efficiencies, or 97.5 per cent, the true total efficiency would be obtained.

By the adoption of this method the following series of formulæ may be used, which make it possible to check back and forth throughout the entire test and in case certain items do not check, it is possible to determine just where the trouble lies, whether in non-representative sampling either for float tests or analyses, or from mistakes in weights or calculations of the same.

In order that the formulæ may be more easily understood and carried in mind, I have used the same letters for designating the same samples, etc., throughout all of the formulæ.

COAL WASHING FORMULAE

L = Loss of Good Coal in the Refuse.
W = Washed Coal Efficiency.
R = Refuse Efficiency.
T = Total Efficiency.
D = per cent Reduction of the Impurity.
M = Percentage of the Impurity actually removed by washing.
a = per cent of Raw Coal. (Always is 100%).
b = per cent of Raw Coal Float.
c = per cent of Raw Coal Sink.
d = per cent of Ash (or Sulphur) in the Raw Coal.
e = per cent of Washed Coal.
f = per cent of Washed Coal Float.
g = per cent of Washed Coal Sink.
h = per cent of Ash (or Sulphur) in the Washed Coal.
i = per cent of Refuse.
j = per cent of Refuse Float.
k = per cent of Refuse Sink.
l = per cent of Ash (or Sulphur) in the Refuse.

Special Note.—It must be remembered that the same specific gravity (permissible) must be used throughout all calculations with these formulæ. Likewise, where analyses are used, they must all be reduced to a dry basis, otherwise the variation in moisture would greatly affect the results.

Formula No. 1.

For determining the value of (L).

$$L=\frac{j\ i}{b}$$

Formula No. 2.

For determining the value of (W).

$$W=100-g.$$

Formula No. 3.

For determining the value of (R).

$$R=100-j.$$

Formula No. 4.

For determining the value of (T).

$$T=\frac{W+R}{2}$$

Formula No. 5.

For determining the value of (D).

$$D=\frac{d-h}{d}$$

Formula No. 6.

For determining the value of (M).

$$M=\frac{d-(eh)}{d}$$

Formula No. 7.

For determining the value of (i).

$$i=\frac{\frac{f}{g}c-b}{\frac{f}{g}k-j}=\frac{fc-gb}{fk-gj}$$

Or, if value of (e) is known, then, $i=100-e$.

Formula No. 8.

For determining the value of (e).

$$e=\frac{\frac{k}{j}b-c}{\frac{k}{j}f-g}=\frac{kb-jc}{kf-jg}.$$

Or, if value of (i) is known, then, $e=100-i$.

Formula No. 9.

For determining the value of (i) from the analysis data.

$$i=\frac{d-h}{l-h}$$

Formula No. 10.

For determining the value of (l).

$$l=\frac{d-he}{i}$$

Formula No. 11.

For determining the value of (h).

$$h=\frac{d-li}{e}$$

If the Float and Sink Tests are all correct, the following formulæ should check.

Formula No. 12.

$$ji+fe=b$$

and

$$ki+ge=c.$$

I have recently written another paper upon this subject in which I went into more detail in the explanation of my method and other coal-washing problems and any wishing to read that paper will find it in the March issue, 1912, of *Mines and Minerals*.

In conclusion, I again wish to emphasize the great need and importance of such data to coal washing. Were a standard universally adopted, and each coal company engineer to make careful tests of all his washeries along the lines specified, and were he to put this data in the form of the standard report and publish it in some one of the engineering journals, the result would be of very great assistance not only to others engaged in that business, but to himself as well. It would make it possible to gather a valuable lot of data together which eventually would show the exact nature of coal to which each type of washing machine was best adapted and this would most certainly prove to be of great and inestimable value to all.

The sampling of the coal and refuse, whether for analyses or float tests, is one of the most important parts of the work. If the samples are not representative, neither the analytical work or the float tests will have any value, nor can it be expected that the work will check out. Too much attention cannot be given this matter and whenever possible, automatic samplers should be used in taking all samples.

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INFLUENCE OF TEMPERATURE ON THE DETERMINATION OF VOLATILE MATTER IN COAL

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In a previous investigation[1] of the causes of discordant results in the determination of volatile matter by the method[2] recommended by the committee on coal analysis, it was found that the principal cause was in the variation of the heat treatment. Further, in working with different types of coal, evidence was obtained that the semi-bituminous coals from the Pocahontas and New River fields of West Virginia were more sensitive to variation in temperature than the bituminous coal from the Pittsburgh bed.

S. W. Parr[3] in his studies of Illinois coal, concludes that the principal variation depends upon the speed with which the heavier or richer volatile constituents are discharged from the mass.

It is essential therefore to further standardize the present method by specifying a definite temperature or range of temperature at which the determination should be made.

In order to provide data on which to base a practical temperature specification that will give the most uniform analytical results with anthracite and the various types of bituminous coal, a series of determinations at temperatures ranging from 750° C. to 1100° C. was made on five different types of coal and one sample of metallurgical coke.

Apparatus

The arrangement of the apparatus is shown in Fig. 1. All determinations, except where otherwise stated, were made in the same 30-gram platinum crucible (A) which was fitted with a special

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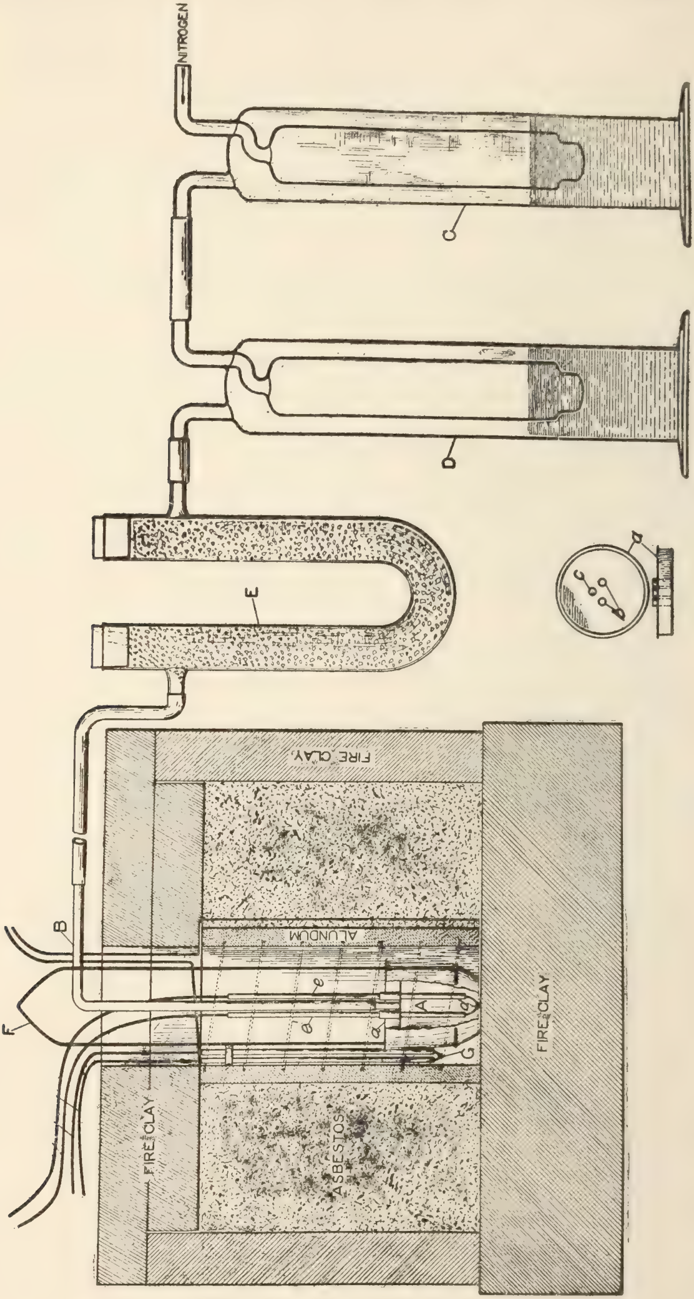
[1] Fieldner and Davis, Journ. Ind. & Eng. Chem. 2, 304 (1910).
[2] Jour. Am. Chem. Soc., 21, 1122 (1899).
[3] Jour. Ind. & Eng. Chem. 4, 352 (1912).


NITROGEN
C
D
E
c
a
FIRE CLAY
ALUNDUM
B
F
e
a
a
A
a
G
FIRE CLAY
ASBESTOS
FIRE CLAY

capsule cover (a). The crucible had a net volume of 25 cc. when the cover was in place. The cover was provided with three platinum tubulures, through two (b) of which were inserted the snugly fitting porcelain tubes that contained the thermocouple leads; to the third tubulure (c) was attached a quartz (B) tube for the introduction of nitrogen. The nitrogen was prepared from sodium nitrate and ammonium chloride by gently heating a mixture of the concentrated solutions according to the method of Knorre.[1]

The description and partial analysis of the samples used are given in the following table:

TABLE I

Description and Partial Analysis of the Air-Dried Samples

Description	Moisture at 105° C.	Ash	Sulphur
Coke	0.20	4.74	0.48
Anthracite, Schuylkill Co., Pa.	0.97	13.18	0.63
Pocahontas Coal, McDowell Co., W. Va.	0.45	8.02	0.62
Pittsburgh Coal, Westmoreland Co., Pa.	0.98	11.23	2.59
Illinois Coal, Vermilion Co., Ill.	3.87	12.48	2.23
Colorado Coal (coking), Las Animas Co., Colo.	1.08	9.05	0.66

The nitrogen was purified by washing with alkaline pyrogallol solution in (C) and potassium bichromate solution in (D) and dried by passing through the U – tube (E) containing granular calcium chloride. The nitrogen as used, contained from 0.3 to 0.4% oxygen.

The temperature of the furnace was controlled by varying the current through a rheostat that was placed in series with the furnace. The furnace temperature was kept constant during each determination by maintaining a constant current through the furnace as shown by an ammeter placed in the circuit. The furnace temperature was measured by thermocouple shown at G. All measurements of the crucible temperature were made with a carefully calibrated platinum-rhodium couple whose junction (d) was in contact with the inside surface of the bot-

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[1] Chem. Industrie — 25, 531 (1902).

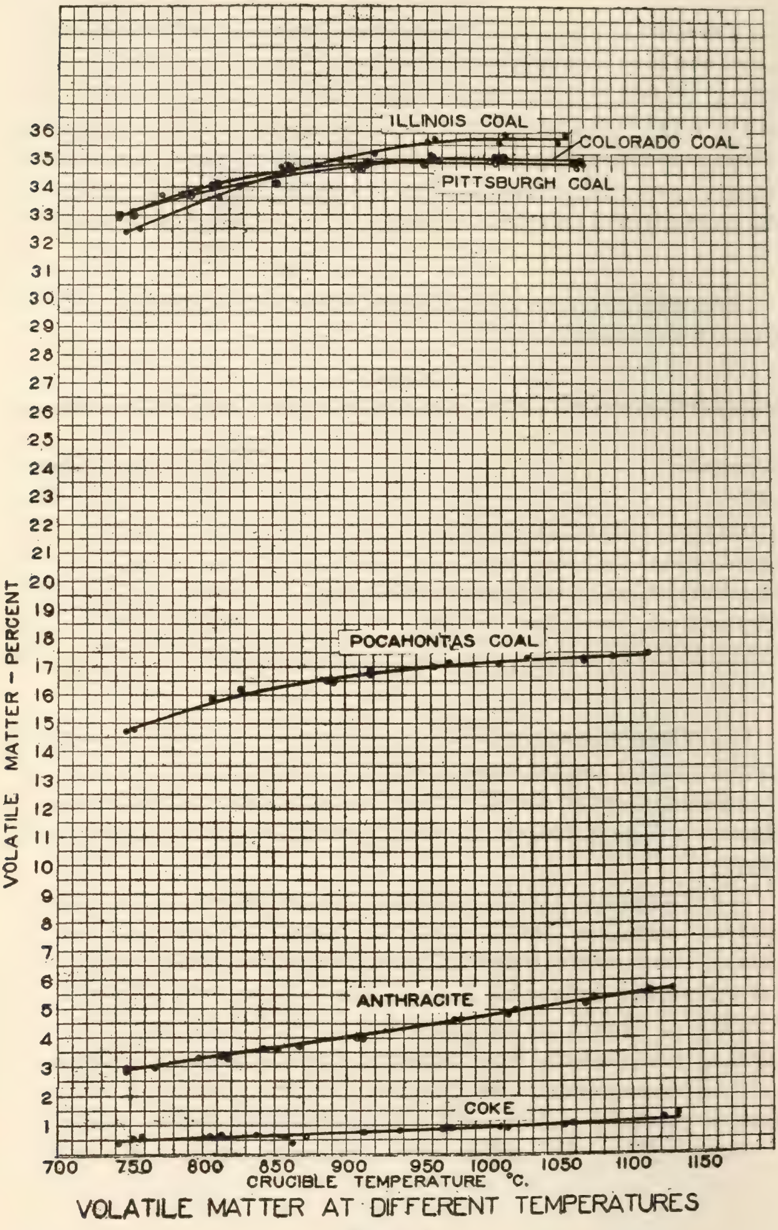

ILLINOIS COAL
COLORADO COAL
PITTSBURGH COAL
POCAHONTAS COAL
ANTHRACITE
COKE
VOLATILE MATTER – PERCENT
36
35
34
33
32
31
30
29
28
27
26
25
24
23
22
21
20
19
18
17
16
15
14
13
12
11
10
9
8
7
6
5
4
3
2
1
700
750
800
850
900
950
1000
1050
1100
1150
CRUCIBLE TEMPERATURE °C.
VOLATILE MATTER AT DIFFERENT TEMPERATURES

Figure 2

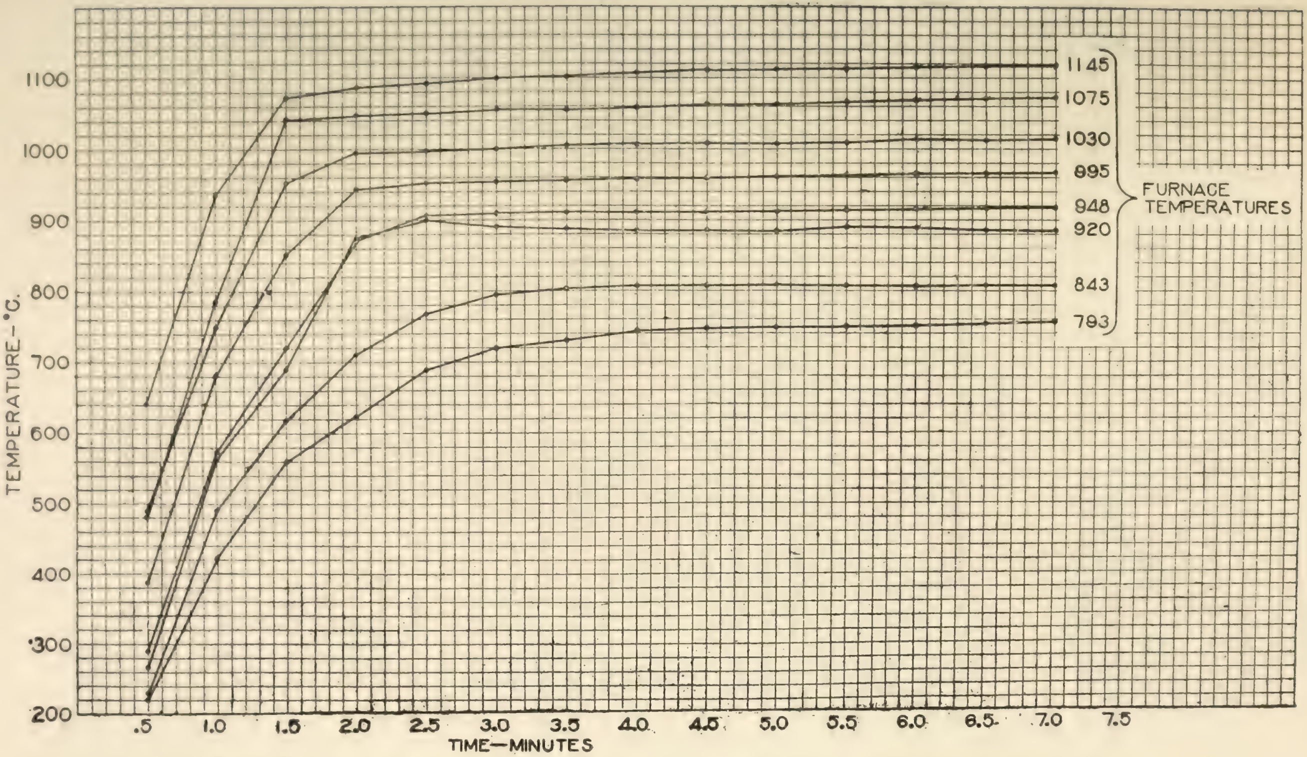


Figure 3—Rise of Temperature in Crucible at different Furnace Temperatures

tom of the crucible. The rather short leads were insulated by porcelain tubes (e) extending through the closely fitting tubulures of the cover. The cold junction was kept at the temperature of melting ice. Means were provided for readily disconnecting the thermocouple at the cold junction, to enable weighing the crucible with cover and thermocouple before and after each determination.

METHOD

The method of procedure in the determination was as follows:

The crucible and cover with attached thermocouple was weighed and one gram of the air-dried sample, pulverized to 60-mesh size, was transferred to the crucible and the cover replaced. The crucible was then placed in a platinum wire support (F) and suspended about 30 centimeters above the furnace opening. The radiant heat from the furnace was cut off by an asbestos board. The quartz tube (B) was then slipped over the projecting platinum tubulure on the cover and a current of nitrogen at the rate of four bubbles per second passed into the crucible for ten minutes. Without interrupting the current of nitrogen, the crucible was then quickly lowered into the hot furnace, the temperature of which had previously been brought to the desired point. This temperature was kept practically constant during the determination; temperature readings were taken at one-half-minute intervals. The rapid evolution of volatile matter began about ten seconds after the crucible was lowered into the furnace and continued for a period of about one minute, the time varying slightly with the furnace temperature. During this period of rapid evolution of volatile matter the current of nitrogen was interrupted by disconnecting the quartz, and platinum tubes to avoid pressure in the crucible; after the rapid evolution had subsided the tube was replaced and the current of nitrogen continued. The time of heating was exactly seven minutes; at the end of the seventh minute the crucible was quickly lifted out of the furnace and allowed to cool without interrupting the current of nitrogen. Meanwhile the thermocouple leads were disconnected and as soon as cool the covered crucible with the thermocouple attached was at once weighed.

The difference in the results of duplicate determinations at the same temperature was usually less than 0.2 per cent. A few duplicate determinations made on the coals high in volatile matter differed about 0.3 per cent. This was probably due to the decomposition of some of the heavy hydrocarbons in the porcelain thermocouple tubes with a consequent deposition of carbon.

In order to determine the extent of any oxidation that might have taken place during the determination either from incomplete expulsion of air or from the small percentages of oxygen in the nitrogen, a number of tests were made by repeating the seven minutes' heating in the furnace after the first loss in weight had been taken and again weighing. The method of procedure in this second heating of the sample was the same as in the first heating. The crucible cover was lifted a few minutes before starting the second experiment in order that no residual nitrogen be left in the crucible from the first heating and thus minimize the task of sweeping out the air in the crucible by the current of nitrogen. The results are given in the following table:

TABLE II

LOSS IN SECOND HEATING

Coal	Mean crucible temperatures during last three minutes C°	Loss in weight in per cent of one-gram sample
Pocahontas	1004	0.15
Pocahontas	1078	0.15
Anthracite	912	0.20
Anthracite	997	0.30
Anthracite	1078	0.35
Coke	757	0.20
Coke	912	0.05
Coke	1020	0.20
Coke	1070	0.25

In one of the above tests a sample of coke was heated for one hour at about 1000° C. then weighed and the determination made by heating seven minutes in an atmosphere of nitrogen. A loss in weight of 0.20 per cent was again obtained. Therefore it would seem that the additional losses in Table II are due to oxidation rather than to further evolution of volatile matter.

The curves in Fig. 2 have not been corrected for the oxidation loss as it is constant, within the experimental error of the determination.

EXPERIMENTAL DATA

The results obtained in the determination of volatile matter at various temperatures according to the above-described method are given in Table III, and graphically illustrated in the curves of Fig. 2.

TABLE III

VOLATILE MATTER AT DIFFERENT TEMPERATURES

O° Temperatures	Coke V.M. in %	Anthracite V.M. in %	Pocahontas V.M. in %	Pittsburgh V.M. in %	Colorado V.M. in %	Illinois V.M. in %
740– 745	0.40			32.86	32.99	
745– 750		2.84 2.96	14.66			32.38
750– 755	0.51		14.80	33.13	33.00	
755– 760	0.56					32.51
765– 770		2.93				
770– 775				33.68		
785– 790				33.69		
790– 795					33.70	
795– 800		3.25		33.89		
805– 810	0.55 0.54		15.85	34.05	34.07	
810– 815	0.60	3.33		34.11		33.60
815– 820		3.23				
825– 830			16.17			33.98
835– 840	0.63					
840– 845		3.57				
850– 855		3.59			34.12	
855– 860	0.53			34.70		34.54
860– 865	0.44			34.75		34.63
865– 870		3.70				
870– 875	0.53					
885– 890			16.49			
890– 895			16.45			
895– 900						34.99
905– 910				34.57		
910– 915	0.68	3.93 4.15			34.62	
915– 920			16.70 16.92	34.75	34.80	

C° Temperatures	Coke V.M. in %	Anthracite V.M .in %	Pocohontas V.M. in %	Pittsburg V.M. in %	Colorado V.M. in %	Illinois V.M. in %
920– 925						35.18
935– 940	0.71					
955– 960					34.93	
960– 965			17.03	35.00	34.94	35.60 35.70
965– 970	0.80			34.92		
970– 975	0.85		17.13			
975– 980		4.63 4.63				
1000–1005				34.87		
1005–1010	0.95		17.10	34.82	34.97	
1010–1015	0.91	4.86			35.02	35.55 35.86
1015–1020		5.00				
1025–1030			17.30			
1050–1055	1.02					35.63
1055–1060	1.02					35.89
1060–1065					34.89	
1065–1070		5.19	17.21 17.33	34.72 34.82	34.97	
1075–1080		5.36				
1085–1090			17.35			
1110–1115		5.60	17.49			
1120–1125	1.22					
1125–1130		5.71				
1130–1135	1.25					

These curves, although too few in number to warrant sweeping conclusions, indicate that discordant results may be expected when volatile matter determination are made in the region below 850 degrees where all the curves have a considerable upward slope. A comparatively slight variation of temperature in this part of the curve will produce appreciable differences in volatile matter. The curves for Colorado, Illinois and Pittsburgh coal are practically horizontal between 950° and 1050° C., while the Pocahontas curve retains a slight upward inclination beyond 950° C.

The curve for anthracite is remarkable in that it is a straight line between 750° C. and 1100° C. This marked upward inclination of the anthracite curve confirms the conclusions of Meade and Attix[1] and Shimer,[2] who noted that anthracite and coke gave more volatile matter when heated over the blast lamp than when heated in the flame of a Bunsen burner.

[1] Jour. Am. Chem. Soc., *21*, 1137 (1899).

[2] Jour. Ind. & Eng. Chem. *1*, 518 (1909).

TABLE IV

VOLATILE MATTER DETERMINED, UNDER VARIOUS CONDITIONS, IN THE ELECTRIC FURNACE AT 1000° C.

Kind of coal	A Special 30 gram crucible. Atmosphere of nitrogen. No kerosene		B Special 30 gram crucible. Atmosphere of nitrogen. 20 drops kerosene		C Special 30 gram crucible. Atmosphere of air. 20 drops of kerosene		D Special 30 gram crucible. Atmosphere of air. No kerosene		E 30 gram crucible. Ordinary cover. Atmosphere of air. No kerosene		F 10 gram crucible, capsule cover. At atmosphere of air. No kerosene	
			0.57									
	0.95		0.53		0.75		1.37		2.45		.92	
Coke	*0.91*	0.93	*0.58*	0.56	*0.60*	0.68	*1.30*	1.33	*2.40*	2.42	*1.12*	1.02
	5.00		4.83		4.83		5.31		6.38		5.00	
Anthracite	*4.86*	4.93	*4.78*	4.80	*4.95*	4.89	*5.43*	5.37	*6.13*	6.25	*5.08*	5.04
	17.10		17.25		17.63		17.62		18.27		17.14	
Pocahontas	*17.30*	17.20	*17.33*	17.29	*17.65*	17.64	*17.65*	17.64	*18.80*	18.53	*17.13*	17.14
			35.29									
	34.82		35.72		35.47		35.20		35.89		34.14	
Pittsburgh	*34.87*	34.84	*35.57*	35.53	*35.75*	35.61	*35.07*	35.13	*35.74*	35.82	*34.73*	34.44
	35.55		36.13		36.43		35.95		37.58		36.37	
Illinois	*35.86*	35.70	*35.83*	35.98	*36.40*	36.41	*36.08*	36.02	*39.15*	38.36	*36.01*	36.19

From the analytical standpoint 1000 degrees C. appears to be the most favorable temperature for the determination of volatile matter. A deviation of twenty-five degrees from this point would not produce a serious change in the yield of volatile matter, except in the case of anthracite, and since this curve is a straight line, the difference in volatile matter for any two consecutive points would be the same for any part of the curve.

In Table IV are given the results of a number of determinations made in the electric furnace, at crucible temperatures within the limits of 1000° and 1030° C. to show the effect of the addition of kerosene, and of the use of the capsule cover with both the 30-gram and 10-gram crucible, as a means of reducing oxidation when making the determination in air. In columns A, B, C, and D the same crucible with the special cover was used as shown in Fig. 1. In column E, an ordinary crucible cover was used on the same 30-gram crucible. In column F a 10-gram crucible with capsule cover was used. This cover was the cap from the bottom of a 20 cc. platinum Gooch crucible. The net volume of the 30-gram crucible with the capsule cover in place was 25 cc. The net volume of the 10-gram crucible with the capsule cover in place was about 5 cc.

The method of procedure of the experiments in columns A and B was the same as previously described. This procedure was slightly modified for the experiments in columns C and D in that no nitrogen was passed into the crucible either before or during the seven-minute period of heating. The quartz tube (B, Fig. 1), however, was kept connected, except during the period of rapid evolution of volatile matter. Just before lifting the crucible out of the furnace the current of dry nitrogen was started and kept flowing into the crucible until it had cooled in order to prevent absorption of moisture by the dry coke. In the experiments in columns E and F the crucible was cooled in a desiccator over calcium chloride.

A comparison of the results given under A, B, C and D indicate little, if any, advantage in the use of kerosene. In the case of coke and anthracite a few drops will aid in reducing oxidation, by expelling the air in the crucible when nitrogen is not used. A few tests made to determine the weight of carbon deposited in the

30-gram crucible from 20 drops of kerosene by heating in nitrogen gave an average carbon deposit on the crucible walls that weighed .0015 grams. The same experiment repeated with the 10-gram crucible with capsule cover gave no deposit. This may partly account for the low results obtained with coke in column B.

A comparison of the results in columns D and E shows that oxidation is materially reduced by the use of a tightly fitting capsule cover in place of the ordinary crucible cover. This oxidation is further reduced by the use of a 10-gram crucible with capsule cover to a point that is in practical agreement with the results obtained in an atmosphere of nitrogen, as shown by a comparison of columns A and F.

TABLE V

A COMPARISON OF THE ELECTRIC FURNACE AND THE NO. 4 MEKER BURNER FOR THE DETERMINATION OF VOLATILE MATTER AT 1000 °C.

Kind of Coal	Electric Furnace 30-gram crucible. Atmosphere of Nitrogen		No. 4 Meker burner. Natural Gas(at an average pressure of 12.5^4 of water). Flame 17 cm. high. Bottom of crucible 5 cm. above burner. 30 gram crucible. Atmosphere of Nitrogen		Differences in Average Values
	% Volatile matter	C.	% Volatile Matter	°C.	
Coke	0.95	1010	1.17	1000	
	0.91	1020	*1.10*	1011	+0.20
	0.93		1.13		
Anthracite	5.00	1019	5.43	1010	
	4.86	1012	*5.28*	1013	+0.42
	4.93		5.35		
Pocahontas	17.10	1010	17.25	1015	
	17.30	1028	*17.36*	1026	+0.08
	17.20		17.28		
Pittsburgh	34.82	1010	34.77	1031	
	34.87	1003	*34.90*	10.27	−0.02
	34.85		34.83		
Illinois	35.55	1012	36.38	992	
	35.86	1014	*36.06*	989	+0.52
	35.70		36.22		

In Table V is given a comparison of the results obtained by the use of the electric furnace and the No. 4 Meker burner in the determination of volatile matter at about 1000° C. There seems to be a tendency to get slightly higher results in the determinations made over the gas burner; this may be due to the more rapid rise

TABLE VI

TEMPERATURE INSIDE CRUCIBLE IN °C.

Atmosphere of Nitrogen (Crucible bottom 3 cm above No. 4 Meker burner, using Pittsburgh natural gas, flame about 18 cm. high.

Time in minutes	.5	1.0	1.5	2.0	2.5	3.0	3.5	4.0	4.5	5.0	5.5	6.0	6.5	7.0	Gas pressure in inches of water
Coke	632	944	982	990	996	999	1005	1008	1008	1010	1011	1011	1010	1010	11.6
Anthracite	632	915	1008	1018	1014	1015	1014	1014	1015	1015	1013	1015	1012	1012	12.5
Pocahontas	592	870	997	1019	1015	1014	1014	1017	1017	1015	1014	1016	1013	1012	12.0
Pittsburgh	582	870	1027	1027	1027	1027	1026	1026	1027	1027	1027	1027	1027	1028	13.0
Illinois	532	870	980	980	980	984	980	980	986	988	989	989	990	990	13.0
Atmosphere of Nitrogen (in the electric furnace)															
Coke	563	870	951	980	990	998	1001	1005	1008	1010	1011	1011	1012	1014	
Anthracite	440	805	935	990	1005	1008	1011	1012	1015	1017	1020	1022	1025	1026	
Pocahontas	490	750	953	995	998	1000	1005	1007	1008	1008	1010	1015	1013	1015	
Pittsburgh	398	652	910	998	998	1000	1001	1003	1006	1008	1009	1014	1016	1016	
Illinois	387	738	964	986	990	998	1000	1005	1008	1009	1012	1014	1016	1017	
Atmosphere of Nitrogen and 20 drops of Kerosene (in the electric furnace)															
															Average furnace Temp. °C.
Coke	532	890	955	978	992	998	1004	1008	1011	1015	1017	1017	1017	1019	1071
Anthracite	418	843	953	992	1001	1008	1010	1015	1017	1018	1019	1019	1020	1024	1072
Pocahontas	430	738	944	982	990	992	997	999	1001	1003	1005	1008	1010	1012	1055
Pittsburgh	461	720	964	985	988	995	998	1003	1005	1007	1009	1012	1014	1016	1053
Illinois	418	729	959	986	993	998	1005	1009	1011	1016	1016	1018	1018	1018	1053
Atmosphere of Nitrogen (in the electric furnace)															
															Average furnace Temp. °C.
Pocahontas	221	423	560	623	689	720	731	744	747	749	750	752	753	756	793
Pocahontas	231	491	619	710	768	795	803	807	808	808	807	807	807	807	843
Pocahontas	268	563	691	875	900	891	888	886	886	885	891	890	887	887	920
Pocahontas	290	573	720	871	906	910	912	912	913	914	915	916	917	918	948
Pocahontas	387	681	852	944	953	954	956	958	960	962	963	965	966	968	995
Pocahontas	490	750	953	995	998	1000	1005	1007	1008	1008	1010	1015	1013	1015	1030
Pocahontas	481	785	1042	1047	1051	1056	1056	1058	1063	1064	1068	1069	1072	1073	1075
Pocahontas	642	935	1074	1087	1094	1099	1101	1108	1111	1112	1113	1115	1117	1118	1145

of temperature that takes place when the crucible is placed in the gas flame. The temperature readings that were taken at one-half-minute intervals during the seven-minute period of heating, for a number of typical determinations, are given in Table VI and Fig. 3.

SUMMARY

Curves showing the influence of temperature on the yield of volatile matter are given for five typical coals and one coke. The character of these curves varies with different types of coal.

Anthracite and semi-bituminous coal suffer a greater change in volatile matter with a given change in temperature than bituminous coal.

1000° C. is recommended as the standard crucible temperature; in the work given in this paper the reference point for temperature measurement was inside the crucible with the junction of the thermocouple in contact with the bottom.

The use of a small crucible, 10 or 15 cc. capacity, with a closely fitting capsule cover reduces oxidation and promotes more uniform results.

Either the electric furnace or the Meker burner may be used, provided the desired temperature can be obtained and properly regulated. However, the electric furnace has the advantage of more accurate regulation.

A COMPARISON OF THE ENGLER AND SAYBOLT VISCOSITIES OF MIXED OILS

By T. T. Gray

Tide Water Oil Company, Bayonne, N. J.

A recent study[1] of the viscosities of mixed lubricating oils has suggested to the writer the desirability of determining, whether, when two oils, one of high and one of low viscosity, are mixed in tenths by weight and the viscosities of the mixtures taken, the curves expressing the results will be similar when different viscosimeters of the same general type are used. The Engler and Saybolt instruments were selected for comparison, and the results obtained plotted.

The Engler results are expressed as specific viscosities, obtained by dividing the time of the flow of the oil, by the time of flow of distilled water at 20° C.; and the Saybolt viscosities as the time of flow of the oil in seconds, using in the first series the old style instrument for engine oils, and in the second and third series, the Saybolt "Standard Universal Viscosimeter." All samples worked on were made from Pennsylvania Crude Petroleum.

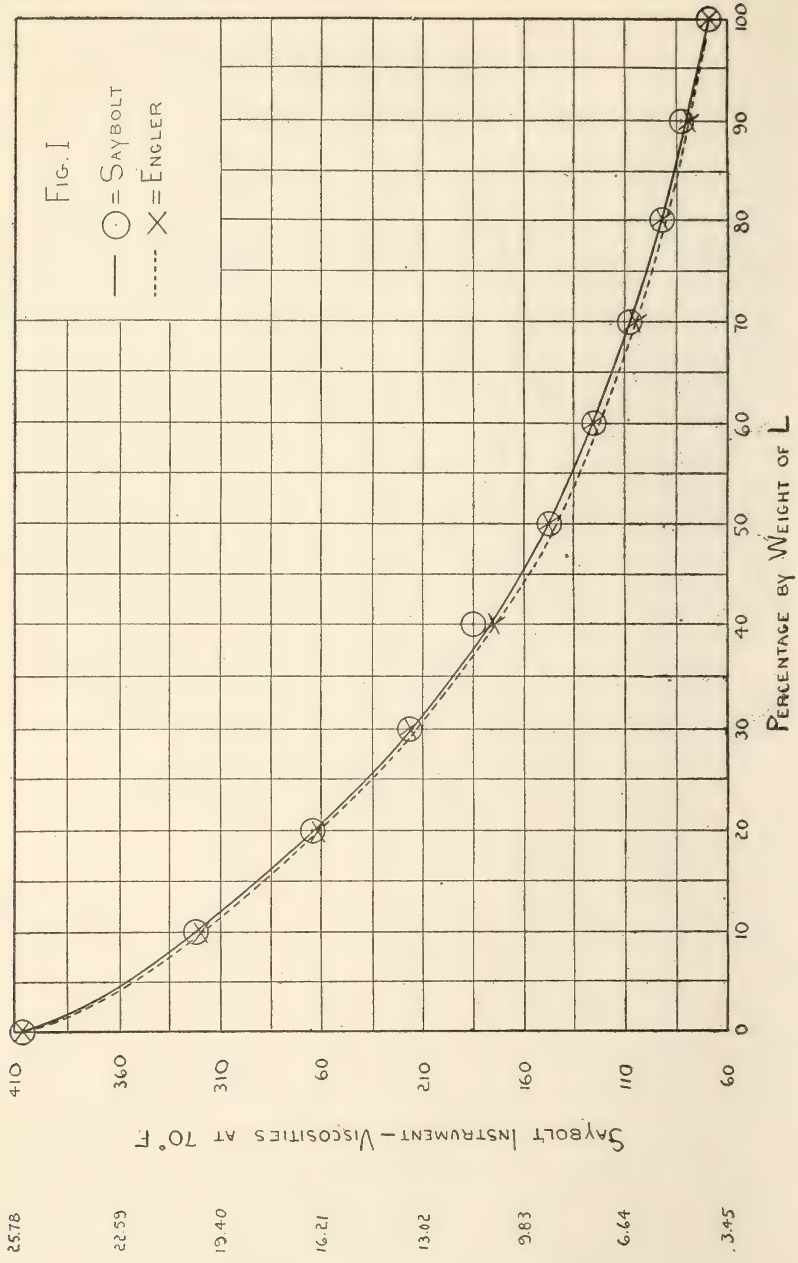
FIRST SERIES

This series was made up of mixture in tenths by weight of a low viscosity lubricating oil ("L") and a high viscosity lubricating oil ("H").

	A Engler Instrument Specific Viscosities at 20° C.	B Saybolt Instrument Viscosities at 70° F.	$\frac{B}{A}$
High Viscosity Oil ("H")	25.56	406	15.88
90% "H," 10% "L"	20.04	321	16.02
80% "H," 20% "L"	16.25	265	16.31
70% "H," 30% "L"	13.37	216	16.16
60% "H," 40% "L"	10.90	185	16.97
50% "H," 50% "L"	9.04	149	16.48
40% "H," 60% "L"	7.68	126	16.41
30% "H," 70% "L"	6.39	108	16.90
20% "H," 80% "L"	5.52	92	16.67
10% "H," 90% "L"	4.67	83	17.77
Low viscosity Oil "L"	4.10	70	17.07

The results are plotted in Fig. 1. (The viscosities as ordinates, and the percentage of "L" as abscissæ.)

Fig. I



○ = Saybolt
X = Engler
Percentage by Weight of L
0
10
20
30
40
50
60
70
80
90
100
Saybolt Instrument — Viscosities at 70° F
410
360
310
60
210
160
110
60
Engler Instrument — Specific Viscosities at 20° C.
25.78
22.59
19.40
16.21
13.02
9.83
6.64
3.45

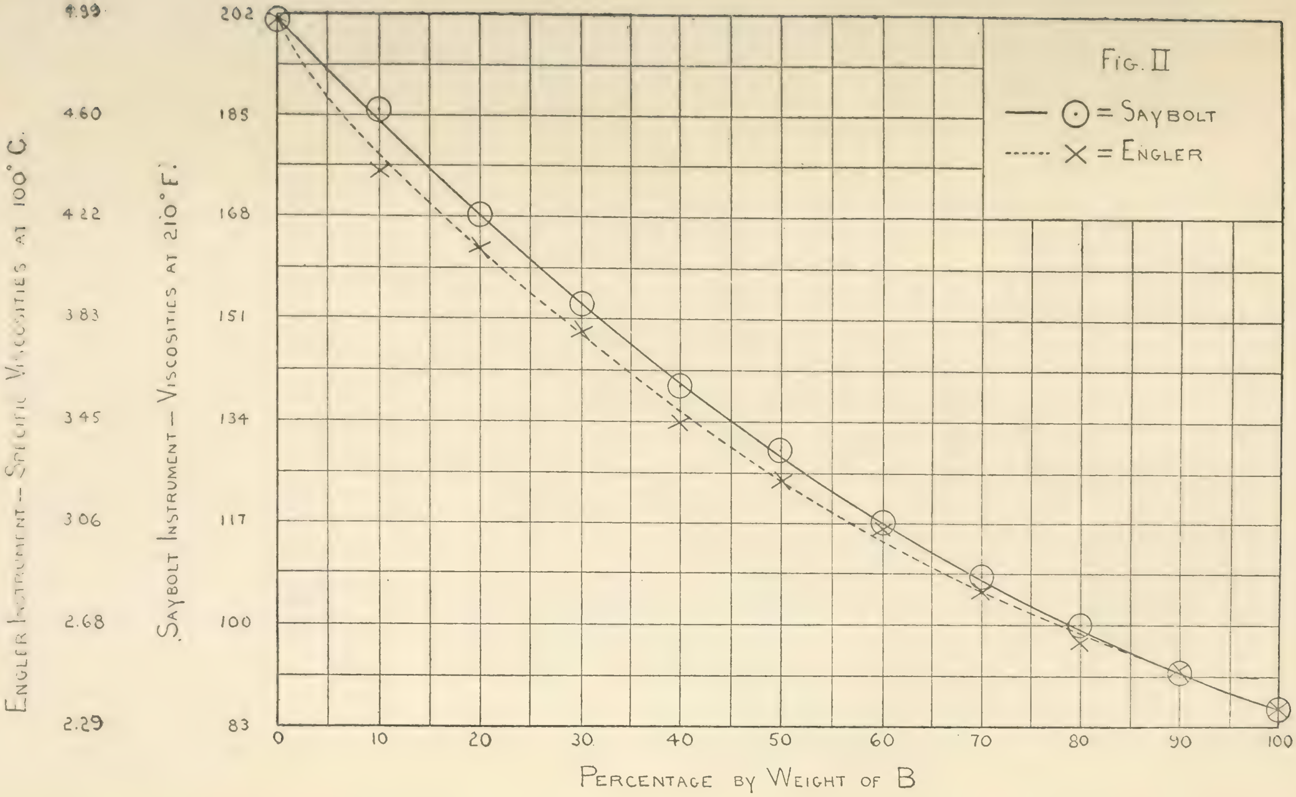


Fig. II
— ⊙ = Saybolt
----- × = Engler
Engler Instrument — Specific Viscosities at 100° C.
4.99
4.60
4.22
3.83
3.45
3.06
2.68
2.29
Saybolt Instrument — Viscosities at 210° F.
202
185
168
151
134
117
100
83
0
10
20
30
40
50
60
70
80
90
100
Percentage by Weight of B

SECOND SERIES

In this case, the samples were prepared from a high viscosity steam refined cylinder oil "S," and a low viscosity Black Oil "B." The mixtures were tested with the following results:

	A Engler Instrument Specific Viscosities at 100° C.	B Standard Universal Instrument Viscosities at 210° F.	$\frac{B}{A}$
Cylinder Stock "S"	4.96	201	40.52
90% "S," 10% "B"	4.39	186	42.37
80% "S," 20% "B"	4.10	168	40.98
70% "S," 30% "B"	3.78	153	40.48
60% "S," 40% "B"	3.43	140	40.82
50% "S," 50% "B"	3.22	129	40.06
40% "S," 60% "B"	3.04	117	38.48
30% "S," 70% "B"	2.80	108	38.57
20% "S," 80% "B"	2.61	100	38.31
10% "S," 90% "B"	2.51	92	36.65
Black Oil "B"	2.37	86	36.29

These results are shown graphically in Figure 2.

THIRD SERIES

The samples of this series were made from two highly refined oils, one of high and one of low viscosity, which are designated ("C") and ("D") respectively.

These mixtures were tested with the following results:

	A Engler Instrument Specific Viscosities at 100° C.	B Standard Universal Instrument Viscosities at 210° F.	$\frac{B}{A}$
High Viscosity Oil "C"	3.86	152	39.38
90% "C," 10% "D"	3.41	131	38.42
80% "C," 20% "D"	3.02	117	38.74
70% "C," 30% "D"	2.75	102	37.09
60% "C," 40% "D"	2.49	91	36.55
50% "C," 50% "D"	2.24	81	36.16
40% "C," 60% "D"	2.02	74	36.63
30% "C," 70% "D"	1.88	67	35.64
20% "C," 80% "D"	1.76	62	35.23
10% "C," 90% "D"	1.63	56	34.36
Low viscosity Oil "D"	1.57	53	33.76

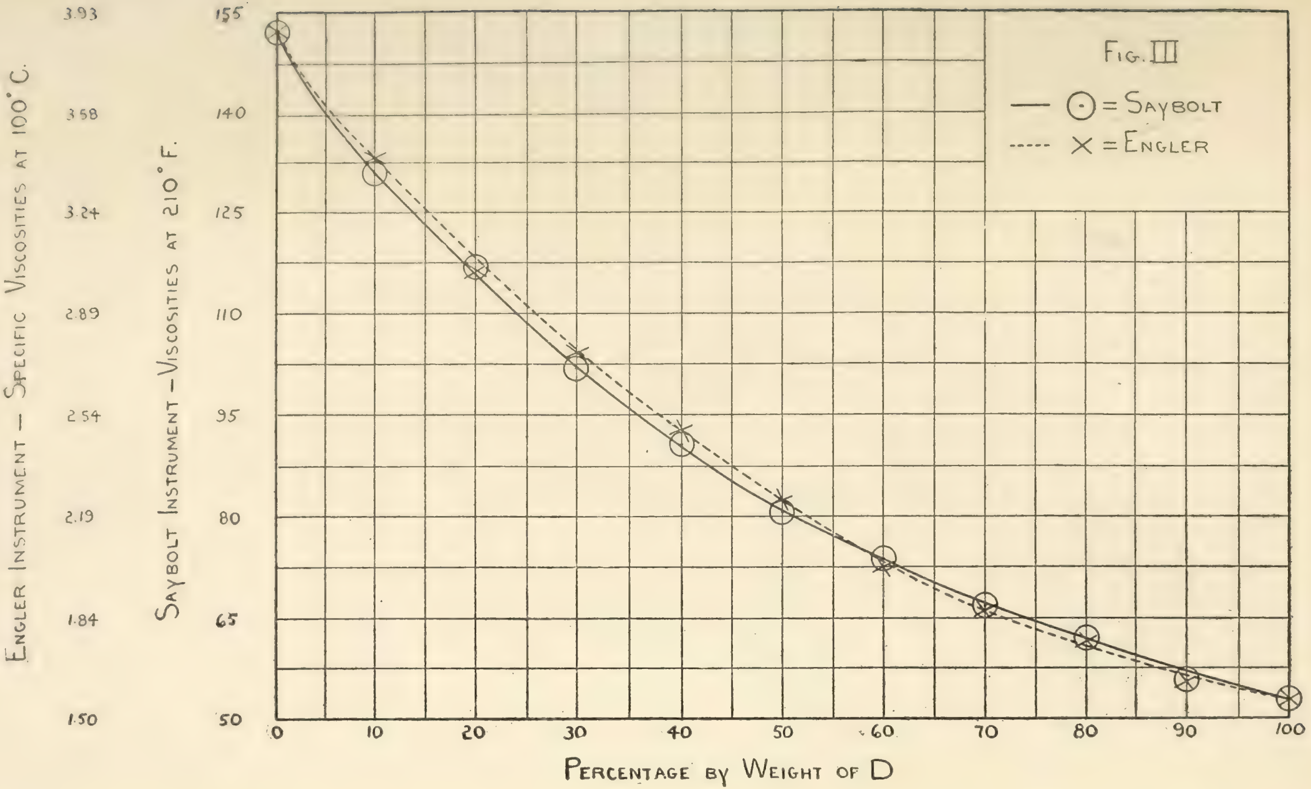


FIG. III
— ⊙ = SAYBOLT
----- X = ENGLER
ENGLER INSTRUMENT — SPECIFIC VISCOSITIES AT 100° C.
3.93
3.58
3.24
2.89
2.54
2.19
1.84
1.50
SAYBOLT INSTRUMENT — VISCOSITIES AT 210° F.
155
140
125
110
95
80
65
50
0
10
20
30
40
50
60
70
80
90
100
PERCENTAGE BY WEIGHT OF D

These results are plotted in Figure 3.

The factors $\frac{B}{A}$ shown in the third columns, were obtained by dividing the Saybolt Viscosities by the Engler Specific Viscosities. From these factors Engler results can readily be converted into Saybolt viscosities, and *vice versa.*

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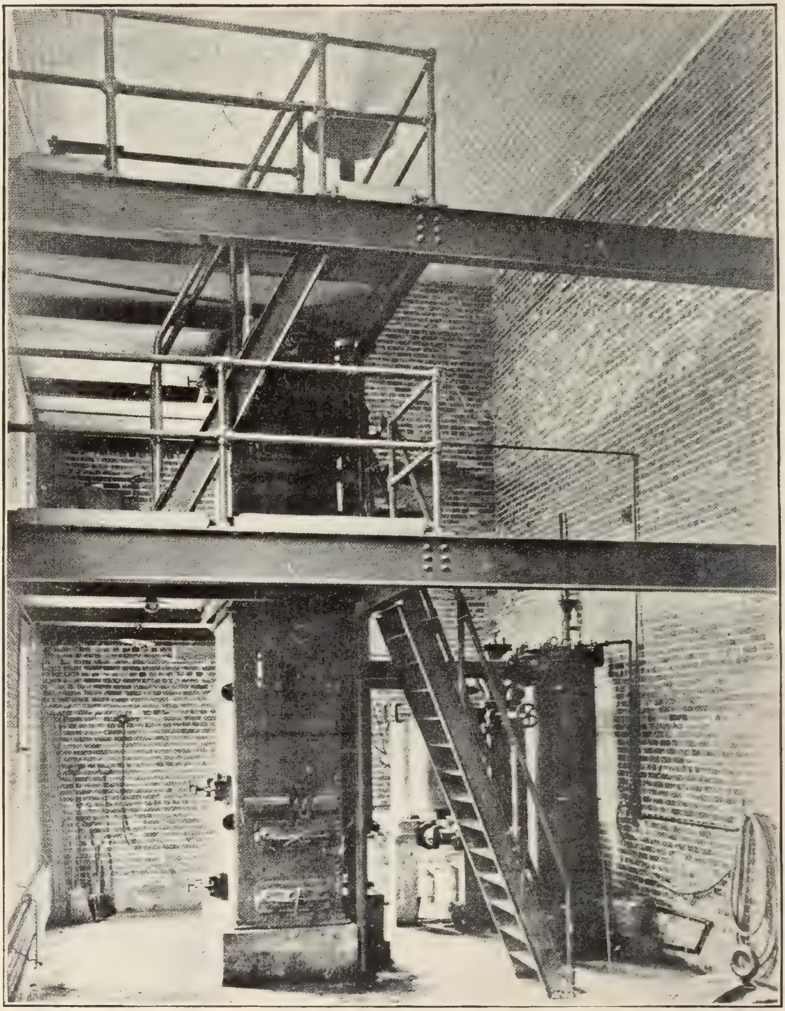
[1] A comparison of the Calculated and Determined Viscosity Numbers (Engler) and Flashing and Burning Points In Oil Mixtures, by H. C. Sherman, T. T. Gray and H. A. Hammerschlag. *Joun. Ind. Eng. Chem.* Vol. 1. No. 1, Jan. 1909.



SIXTY HORSE POWER KÖRTING PEAT GAS PRODUCER AT GOVERNMENT FUEL TESTING STATION, OTTAWA, CANADA.

THE PRODUCTION AND UTILIZATION OF PEAT FOR POWER PURPOSES WITH SPECIAL REFERENCE TO THE KORTING PEAT PRODUCER-GAS POWER PLANT INSTALLED AT THE FUEL TESTING STATION OF THE DEPARTMENT OF MINES, OTTAWA, CANADA

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The active interest taken by the Canadian Government in assisting the exploitation of peat bogs and the establishment of a peat fuel industry on a firm commercial basis was prompted by the dependence of the central provinces of Canada on some foreign source for their fuel supply.

This dependence is due to an absence of coal measures; the rapidly decreasing supply of wood, which can no longer meet the demands for industrial and domestic purposes; and finally, the prohibitive cost of hauling coal from our own mines — situated in the extreme east and west — to distributive points in these provinces.

But while this portion of Canada is devoid of bituminous or anthracite coal measures, large areas are covered with peat bogs which contain peat of excellent quality, eminently suitable for the manufacture of peat fuel. If all the peat bogs possessing the requisite qualities were exploited for the manufacture of fuel, the demand for both domestic and industrial purposes could be met for years to come, and the Provinces rendered independent of outside sources for a fuel supply.

The assistance rendered the development and exploitation of these peat bogs by the establishment of a demonstration peat fuel manufacturing plant erected on a bog acquired by the Government, and the installation of a peat producer-gas power plant at the

Fuel Testing Station, Ottawa, has already borne fruit, by awakening general interest in the possibilities of utilizing peat fuel for power and domestic purposes and since these demonstration plants erected by the Government were purchased — after exhaustive investigation by competent engineers — they may be regarded as examples of the most successful and economic methods for the manufacture of peat fuel, and its utilization for power purposes.

The experimental work done by the Canadian Government has led to most gratifying results, especially in the case of the producer-gas power plant, where much original work — involving the complete reconstruction of the plant — had to be done before it could be pronounced a success. For this reason, a description of the process employed for the manufacture of air-dried, machine peat fuel, and its utilization for power purposes, in what I believe to be the only producer-gas plant on this continent especially designed for, and successfully using, peat fuel, may not be without interest.

MANUFACTURE OF AIR-DRIED MACHINE PEAT

The raw peat contained in the average well-drained bog contains from 85-90 per cent of water. Before the peat taken from the bog is suitable for burning, the greater portion of the water must be removed.

Many processes have been invented for drying peat and manufacturing it into a suitable fuel; but the majority of these processses have depended upon drying by the application of artificial heat, and for the mechanical expulsion of the water by means of powerful presses. Such processes are not economic, as costly and exhaustive experimentation has demonstrated.

At the present time the only economic and successful process for manufacturing peat fuel is that which mechanically treats the raw peat, and utilizes the sun's heat for drying. This process is employed at the Government peat plant, Ottawa.

The raw peat excavated from the bog is fed into a pulping mill, which consists of a series of revolving knives, rotating against fixed knives. By means of the mechanical treatment to which the peat is subjected in the mill, the peat from the different layers of the bog is mixed, and the cell walls broken up.

By thoroughly mixing the peat from the different layers in the bog, a homogeneous peat mass results, and the destruction or breaking up of the cell walls enables the contained moisture to evaporate more easily. The quality of the fuel depends on the thoroughness with which these processes are carried out.

After thorough pulping, the peat is taken to the drying field, where it is spread out, and cut up into blocks of suitable size. The surfaces of the peat blocks are now exposed to the wind, and the sun's heat; and when drying on the exposed side has proceeded far enough, the blocks are turned over, thus exposing the under surface to the sun and wind also.

The following are a few of the advantages resulting from the the pulping of the raw peat:

(1) Increase in density, and greater resistance to breakage.

(2) The formation of a gelatinous skin over the surfaces of the peat, which renders it practically waterproof, and prevents the absorption of moisture from without. The usual explanation advanced for this phenomenon is that the skin swells during damp or rainy weather, and closes its pores, which prevents the moisture from entering the interior. In dry weather, these pores open again and the drying process continues at practically the same content of moisture which the peat had before the rain.

(3) Loss in manufacture — due to fines, broken blocks, etc.— is reduced to a minimum.

In addition to the above advantages resulting from this method of manufacturing peat fuel, it has been demonstrated in our manufacturing operations — covering a period of over two seasons, during which time over 3000 tons of peat fuel were manufactured — that the fuel can stand handling and long transportation without breaking, and indefinite storage under cover.

Over 15 tons of peat fuel were piled in the open air, exposed to rain, snow, and severe freezing during the autumn, winter, and spring, at the end of this time, examination showed that no appreciable deterioration, of its physical properties, or heating value, had taken place.

MOISTURE

By open-air drying the moisture content is reduced to 25–30 per cent, and it does not appear to be feasible or economical to reduce the moisture content further. Air-dried machined peat, when ready for the market, for domestic or industrial uses, is, therefore, assumed to contain from 25–30 per cent moisture.

DISADVANTAGES OF THE PROCESS, AS CARRIED ON AT THE GOVERNMENT PLANT

The peat supplied to the peat machine (pulping mill) is dug entirely by hand. This entails the labor of seven men. For filling the cars, one man is employed, while for spreading and cutting the peat three more men are necessary. This number of laborers — together with one engineer, and two boys—brings the total up to 13.

Under the existing condition of the high cost of labor, this hand labor method cannot be employed for the manufacture of a fuel fuel sufficiently cheap to stand the cost of considerable transportation, and still compete with coal on the open market. The estimated cost of one ton (2000 lbs.) of peat fuel, containing 25 per cent moisture, I have estimated to be $2.00, — including interest on capital, sinking fund, repairs, and a small profit of about 15 cents per ton. But this only holds true when the plant is operated at full capacity, namely, 30 tons per day, for a period of about 100 days.

To overcome the disadvantages due to labor conditions, we have recommended that the process be carried out by automatic machinery, as far as possible. The excavation and spreading — the two items involving the largest amount of labor — should be performed automatically. In addition, the output should be increased, and the plant worked night and day; in order to further reduce the overhead charges.

These recommendations have been taken advantage of by a company who are now installing a plant of large capacity, in which hand labor will be largely replaced by mechanical appliances.

GASIFICATION OF PEAT FUEL

By far the most extensive field for the successful utilization of peat in Canada, is in my opinion, offered by its conversion into power — electrical or otherwise — through the medium of producer-gas power plants. On account of the large volume of peat fuel, and its low-heating value, it cannot be recommended for steam-raising purposes.

The high percentage of nitrogen contained in the Canadian peat bogs so far examined, peculiarly adapts the peat for gasification in plants designed for ammonia recovery. And since the imports of artificial fertilizers are annually increasing, the recovery of the nitrogen in the peat, as ammonium sulphate, is not only most important, but should be attempted wherever it is possible to do so economically.

TYPES OF PRODUCERS

There are two principal methods at present employed for the utilization of peat in gas producers:

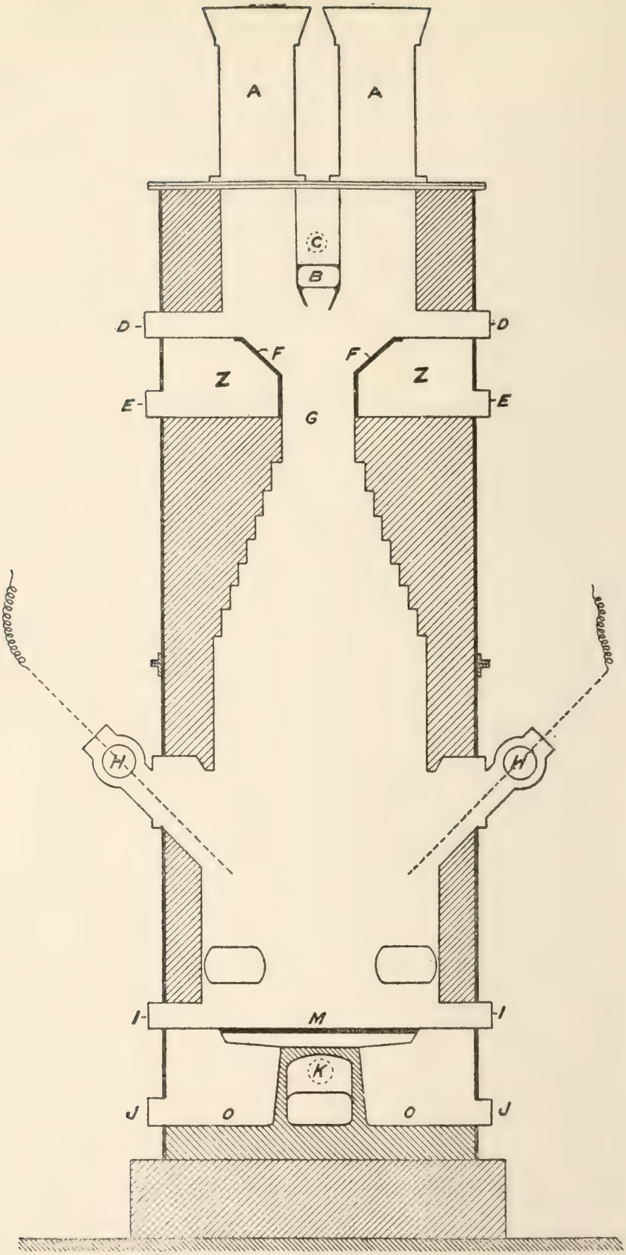
(1) The direct conversion of the heat energy of peat into work by its gasification in a producer not constructed for recovery of by-products; hence a producer designed to burn the fuel with the highest possible thermal efficiency.

(2) The conversion of the heat energy of peat into useful work by its gasification in a producer designed for the recovery of the commercial by-products. The production of power gas might, in this case, be termed the by-products.

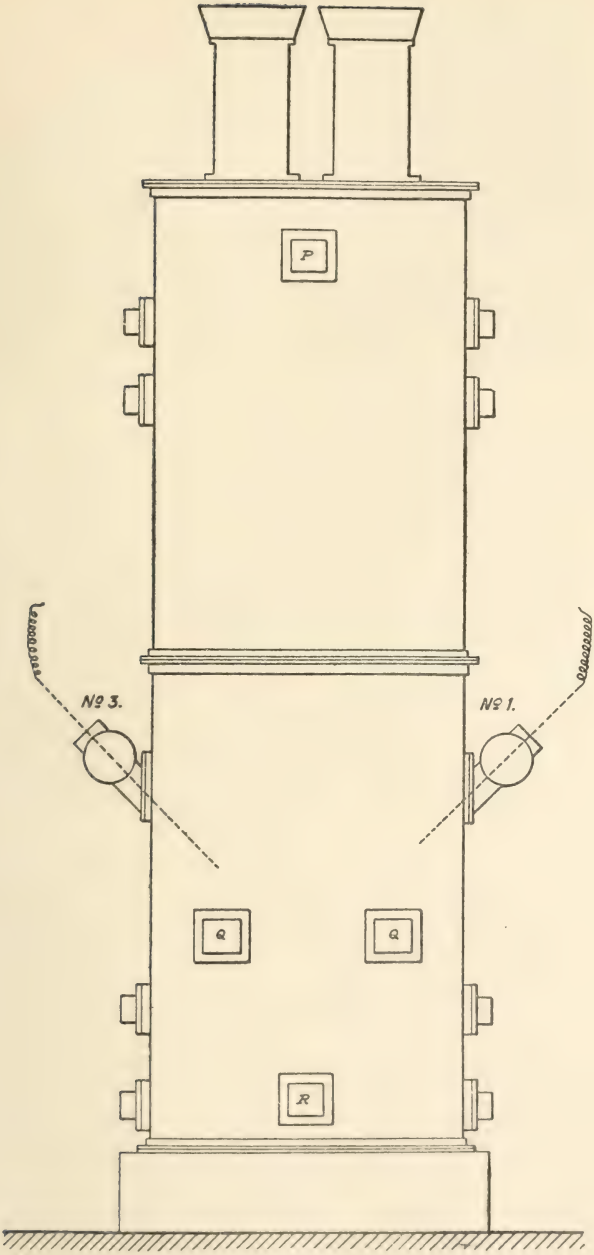
The first type — that designed for the direct conversion of the heat energy of peat into useful work, where the principal aim is the production of power gas with the highest possible thermal efficiency — is the one most commonly in use. To this type belongs the Körting peat gas producer erected at the Fuel Testing Station, Ottawa.

In designing this producer, the makers aimed at the complete gasification of all the combustible components in the peat; no attempt being made to recover any of the by-products, but rather to destroy them.

The small percentage of fixed carbon, large amount of volatile matter, and the comparatively large amount of moisture contained in the peat it is desired to burn in the producer, renders this fuel more or less difficult to use successfully in producers designed for either bituminous coals, or lignite, although some of the properties of lignite closely resemble those of peat. Peat, moreover, is a very poor conductor of heat, and this negative property, to-



Sectional elevation through A-A.
FIG. 2.—Körting Peat Gas Producer.



Rear Elevation.
FIG. 2.—Körting Peat Gas Producer.

gether with the high moisture content, tends to confine the combustion in a down-draft producer to the peat near the lining. Hence it is impossible to maintain the fuel level to insure the required depth of hot peat coke through which to pass the volatile matter and moisture driven off at the top, or, to supply the requisite quantity of gas per square foot of grate surface.

This can be partially overcome by introducing a supply of preheated air into the centre of the fuel bed: thus increasing the surface of combustion.

KORTING PEAT GAS PRODUCER

The difficulties enumerated above are overcome in the producer designed by the Körting Brothers. In this producer two combustion zones are employed: one situated near the top, and the other near the bottom of the producer. The function of the top zone is to distill off the volatile matter, and to evaporate the moisture from the peat. As a result of this operation the peat is coked, and this coke is then fed to the lower zone. The gases, hydrocarbon vapors, and moisture evolved in the upper zone, are now caused to pass through the hot peat coke in the lower zone.

It was expected that these products of the distillation of the peat in the upper zone, by being passed through the incandescent peat coke of the lower zone, would result in a producer gas free from tar.

In practice, however, this has not been realized in the experiments conducted with the Körting producer. A certain quantity of tar was always found to leave the producer with the gas, and required to be separated out by a suitable cleaning system.

DESCRIPTION OF PRODUCER

The producer, as originally erected, is shown in section and elevation, in Figs. 1, 2 and 3, and in perspective in Plate 1. It consists of a rectangular steel shell with the following outside dimensions: 2′-9″ x 5′-2″, in horizontal section, and 15′-0″ high, from floor level to top of charging hoppers. For cleaning fires and removing ashes, 12 doors are provided: 4 on each side, and 4 on the back. These doors are shown on Plate 1.

Figure 2 shows the two combustion zones F-F at the top, and

M at the bottom. F-F represents inclined grate bars: and D-D doors for poking and cleaning the fires resting on the grates F-F. The ashes resulting from the combustion of the fuel on these grates drop into the chambers Z-Z, and are removed from the doors E-E. The gases evolved at this zone are drawn off through the chamber B. The dust and tar which are caught in this chamber are removed through door P on the back of the producer, as shown in Fig. 2. A-A represents charging hoppers.

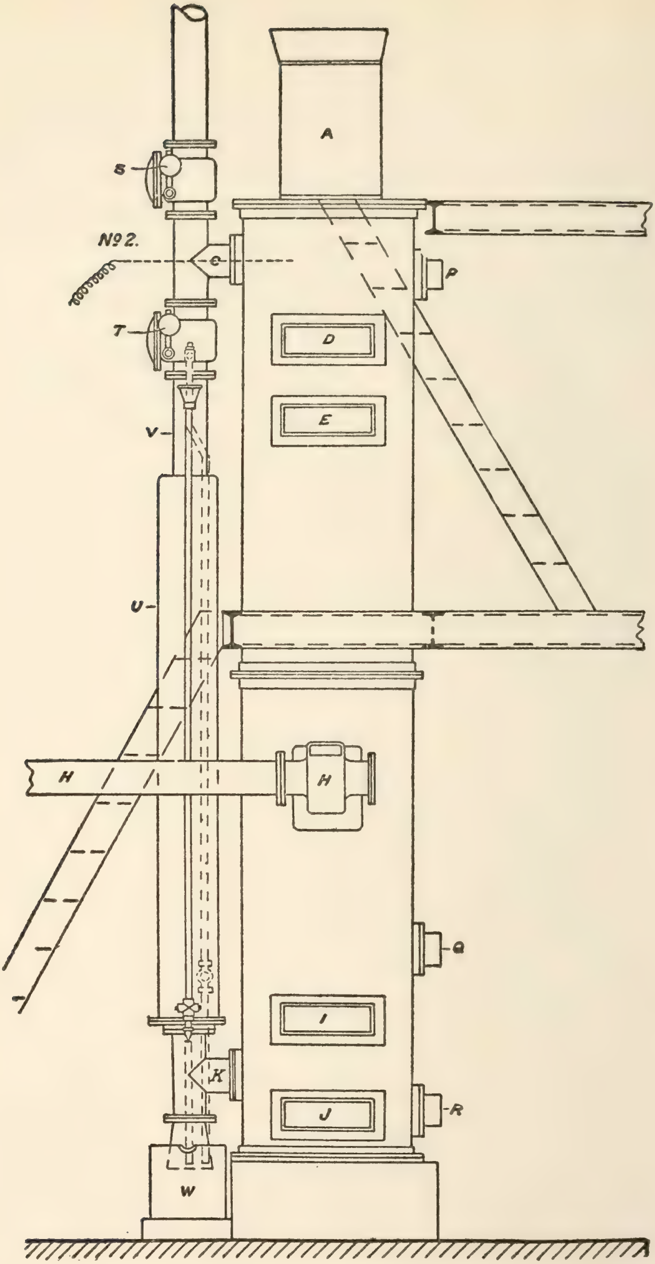
M, on Fig. 1, represents the grate bars of the lower zone, and I-I the doors through which the fires of the lower zone are poked and cleaned. The fire above the grates of the lower zone is poked through the doors Q-Q, as shown in Figs. 2 and 3. The ashes resulting from the complete combustion of the fuel in this zone fall through the grate bars into the chambers O-O, and are removed through the doors J-J.

The products of combustion in the upper zone are drawn off through the chamber B, through the pipe C, and vertical pipe V, shown in Fig. 3, to chamber K, Fig. 1; when they pass finally through the fuel of the lower zone to the two gas off-takes H-H, Fig. 1.

S, on Fig. 3, at the top of vertical pipe V, is a damper which is opened to allow the gases resulting from the combustion of the peat to pass into the atmosphere — when the producer is standing idle. The damper T — also shown in Fig. 4 — is closed while the producer is standing idle; but it is opened and the damper S closed when the producer is in operation. These two dampers are provided with cover plates, which can be readily removed when it is necessary to remove from the damper chambers any material which has collected therein. The vertical gas pipe V, is cooled by means of cold water continually circulating in the jacket U. The cooling water enters at the bottom of this jacket, and overflows at the top to a water seal W, which covers the open bottom of the vertical pipe V.

The ashes which fall through the grate bars M into the gas chamber K — shown in section in Fig. 1 — are removed through door R shown in Figs. 2 and 3.

The off-takes H-H, shown in Fig. 1 are provided with caps held firmly in place by clamps, which may be removed for the purpose



Side Elevation.
FIG. 3.—Korting Peat Gas Producer.

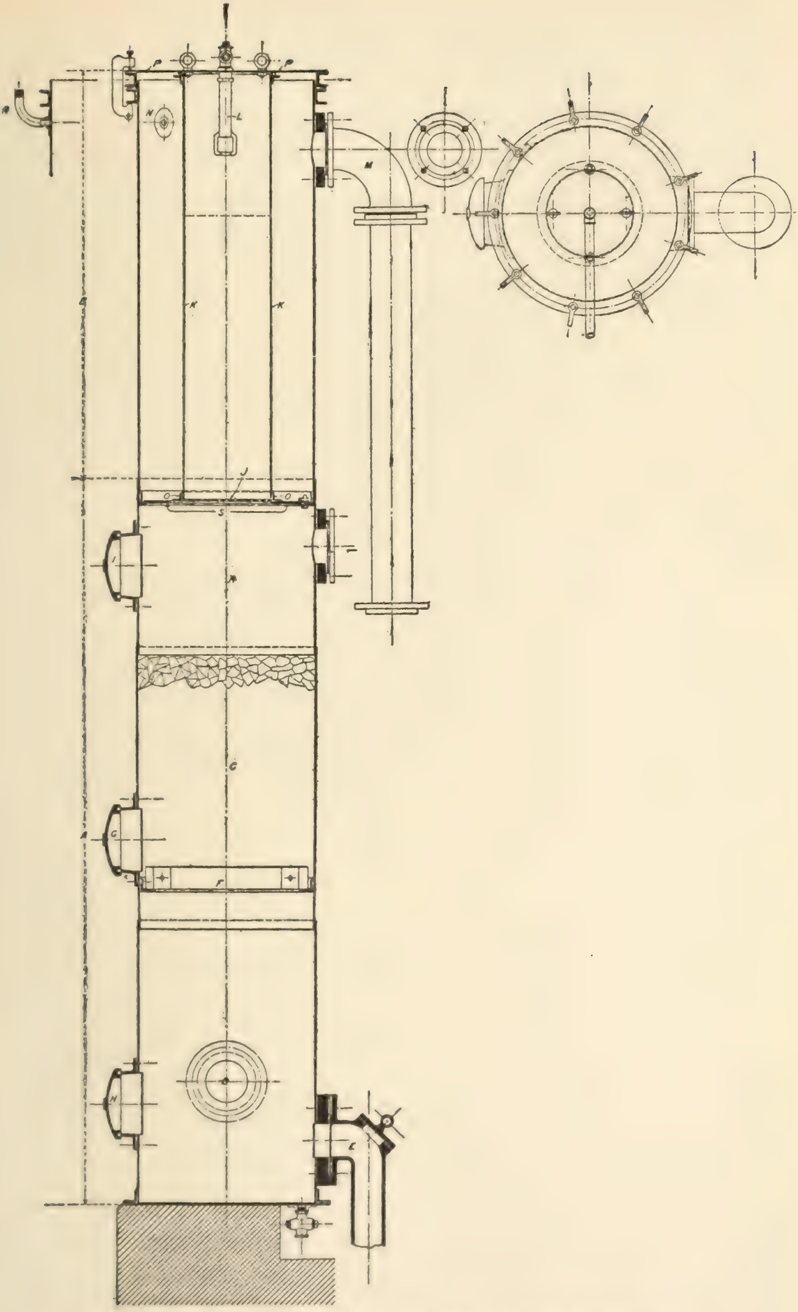


Figure 4. Korting Improved Wet Coke Scrubber

of inspecting the interior of the producer, at this point, or, for the purpose of removing any dust or tarry matter which may collect in the gas chamber.

For the purpose of regulating the amount of air entering the producer at the top and bottom zones, two adjustable air-openings — not shown in the figures — are provided on each of the doors E-E and J-J.

PRINCIPLE OF OPERATION

When the producer is in the proper condition for operation, that portion between the lower grate bars M and the grates F-F (Fig. 1) of the upper zone, is completely filled with peat coke, *i.e.*, peat free from moisture and volatile matter. That portion of the producer between the upper grate level and tops of hoppers A-A, is filled with peat fuel. The function of the upper zone consists in driving the moisture and volatile matter from the peat which supplies the lower zone. To prevent, as far as possible, the products of combustion of the upper zone from being drawn by the suction of the gas engine straight down through the producer and out through the off-takes H-H, instead of being drawn out through C and then having to pass up through the incandescent fuel in the lower zone before reaching H, the construction of the fire brick lining is made as shown in Fig. 1, a contracted neck G being made just below the upper zone.

With this construction it was supposed that the resistance to the passage of the gas through the neck filled with peat coke would be great enough to prevent the gas from being drawn down through the coke to the off-takes — instead of through the chamber B and pipes C and V, Figs. 1 and 3, and chamber K, and then up through the incandescent carbon to the two gas off-takes H-H. This construction, however, did not prove entirely satisfactory, since a portion of the gases evolved in the upper zone passed directly down through the contracted neck to the off-takes, and consequently left the producer with the original moisture and tarry matter driven off in the upper zone.

To overcome this difficulty the construction of the producer was changed — this alteration will be described later.

The combustion taking place at the upper zone should be just sufficient to supply the heat necessary to evaporate the moisture and drive off the volatile matter contained in the peat. The gaseous products, *viz.*, water vapor, tarry vapors, carbon monoxide, carbon dioxide, and a small percentage of gaseous hydrocarbons, in the form of stable gases, resulting from the combustion taking place in the upper zone, are drawn off, as explained previously, through the chamber B (Fig. 1), and down through the water-cooled pipe V (Fig. 3), to the gas chamber K, situated under the fires of the lower zone. Some of the tarry vapors, and probably some moisture are condensed on the water-cooled surface of pipe V and drop to the bottom of the water seal W (Fig. 3): from which the tar can be readily removed. That portion of the moisture and tarry vapors which escapes condensation in passing down through the water-cooled pipe V, is drawn up through the incandescent peat-coke of the lower zone, and through the gas off-takes.

A part of the moisture is decomposed by reaction with the hot carbon, forming free hydrogen carbon monoxide and carbon dioxide. Part of the carbon dioxide is reduced to carbon monoxide, and some of the tarry vapors are decomposed — forming permanent combustible gases.

The following analysis shows the composition of a sample taken from the gases evolved in the upper zone:

CO_2	15.3 per cent by volume
CO	7.2 per cent by volume
O_2	3.2 per cent by volume
C_2H_4	0.7 per cent by volume

The residue being chiefly nitrogen, the gases would also contain water and hydrocarbon vapors, which would condense in the sample bottle before analysis.

The successful operation of this producer depends, as previously stated, on the quality of the fuel fed to the lower zone and the passage of the gases evolved at the upper zone through the bed of incandescent peat coke of the lower zone before leaving the producer. Partially coked peat on the one hand, and the passage of the upper zone gases down through the fuel to the off-takes, on the other hand, or both of these together, will result in a final gas heavily charged with tarry matter; since any volatile matter contained in the fuel fed to the lower zone cannot possibly be

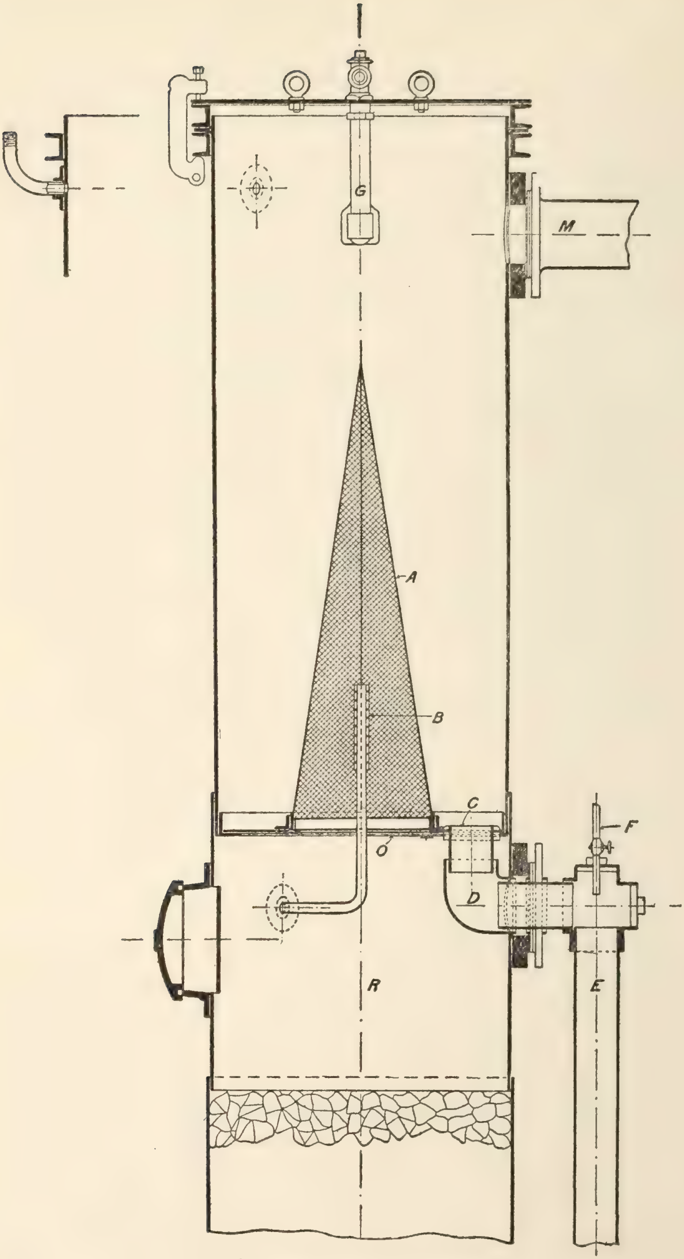


FIG. 6.—Newly devised tar separator.

destroyed or cracked, inasmuch as this volatile matter will be distilled by the heat of the lower zone and pass off without reacting with the hot carbon, or burning. The same condition prevails with regard to the passage of the gases and tarry vapors evolved at the upper zone down through the contracted neck and fuel to the off-takes; since these gases, moisture, and tarry vapors do not come into contact with the hot carbon, hence leave the producer practically unchanged.

With the producer as originally constructed, as shown in section on Fig. 1, both these conditions prevailed to a certain extent, with the result that the cleaning system failed to separate the tar from the gas, consequently the valves and piston of the engine, in a short time, became clogged — necessitating cleaning.

ALTERATIONS TO KORTING PRODUCER

After considerable experimentation to overcome this difficulty — all of which proved futile, I made certain suggestions to the Canadian representative of the Körting Brothers regarding modifications to the producer, the adoption of which, in my opinion, would greatly improve its operation. As a result of these suggestions the Körting Brothers experimented with several tons of peat sent from the Government bog, and finally altered the shape of the lining and made other changes which will be described later. The alterations to the producer are shown on Plate 2.

The principal change in the design is the lengthening of the contracted neck, which ensures greater resistance to the passage of the gases down through the fuel bed. The grates of the upper zone were made with greater air spaces, while those of the lower zone were so designed that the gases passing through the hot fuel of this zone would be deflected towards the off-takes.

These changes tended to greatly improve the operation of the producer; but the gases delivered to the engine still contained so much tar that the piston and cylinder soon became covered with a deposit of tar, which had to be removed.

The grates of the lower zone — after the producer had been in operation some time — became clogged with ashes and clinkers over the openings admitting the upper zone gases, and this upset the operation, forcing the gases to pass directly down through the narrow neck and fuel to the gas off-takes. The construction of these grates covering the gas chamber, previously mentioned, made it impossible to clear this portion of the grates while the plant was in operation — hence such grates are impracticable. The grates used with the producer, as formerly constructed, were made with larger and more simple openings, which permitted ashes to fall through easily into the gas chamber, thus preventing clogging. These grates were substituted for the newly designed ones, and proved more efficient in every respect.

GAS-CLEANING SYSTEM

The gas-cleaning system consists of a wet coke scrubber, tar filter, and dry scrubber. The wet coke scrubber with the improvements incorporated at the time the producer was altered, is shown in Fig. 4. The cleaning system is shown in section in Fig. 5, as it was originally constructed, with the exception of the tar filter, which was added as an extra precaution against tar entering the engine.

On Fig. 4 the addition to the original scrubber is designated by B, and consists of a cylinder about five feet long, rivetted to the top of the old scrubber, which is designated by A. The cap P-P, is fastened to the top of this cylinder by clamps. Inside this cylinder there is placed another cylinder of smaller diameter (K-K) which fits firmly between the cover-plate P-P and the diaphragm O-O. This cylinder is perforated with small holes for about two-thirds of its height. The lower end rests on a perforated plate J. The gas is cooled by the water spray L which also washes off matter sticking to the inside of the cylinder.

The lower portion of the scrubber A is filled with coke as shown at C.

TAR FILTER

This filter is composed of a number of staggered baffle plates around which the gases pass before passing finally through four perforated metal plates. The baffle and perforated plates are washed by sprays of hot water. The hot water for spraying is obtained from the return cooling water of the gas engine.

DRY SCRUBBER

From the tar filter the gas passes through a dry scrubber which consists of a closed cylinder filled with wood fibre.

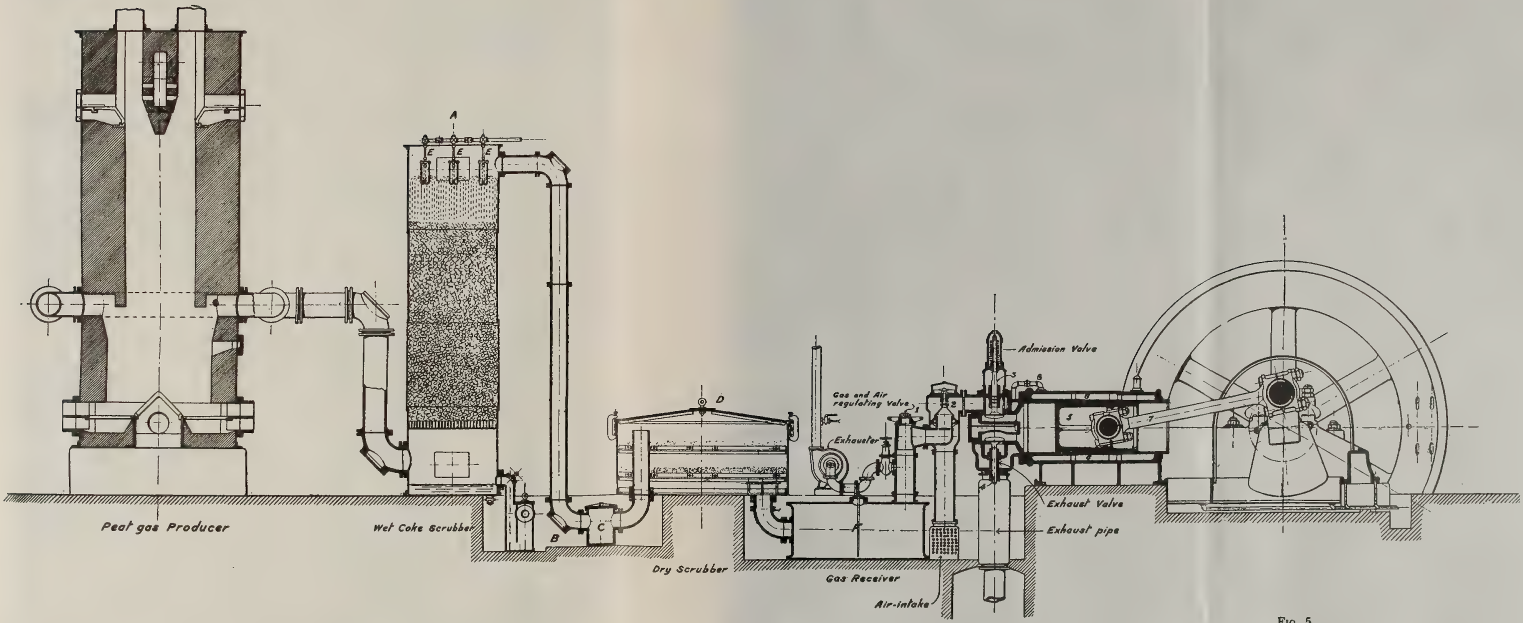


Fig. 5.
IDEAL SECTION
OF
KÖRTING PEAT PRODUCER—GAS PLANT.

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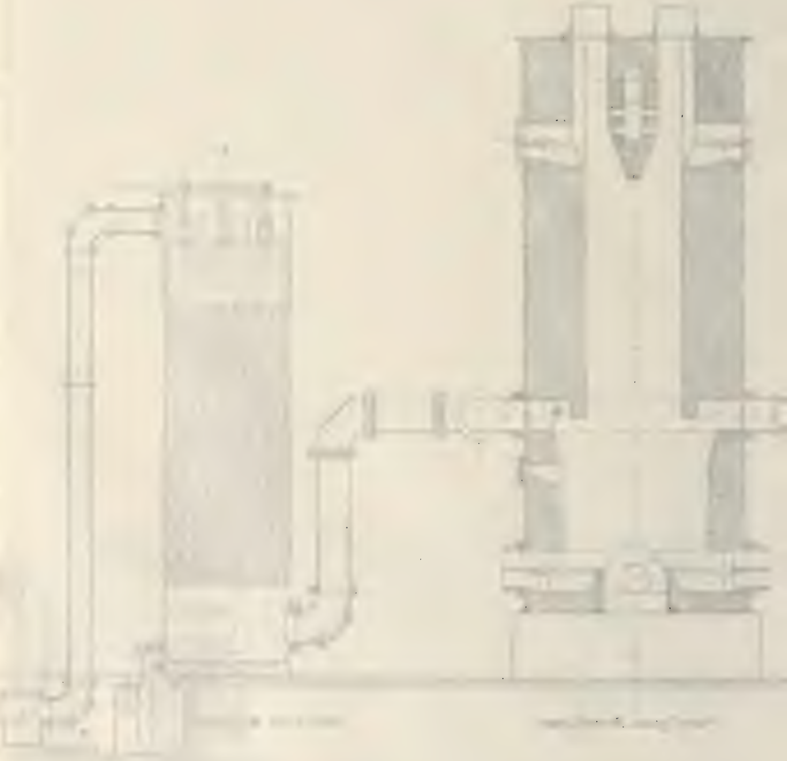


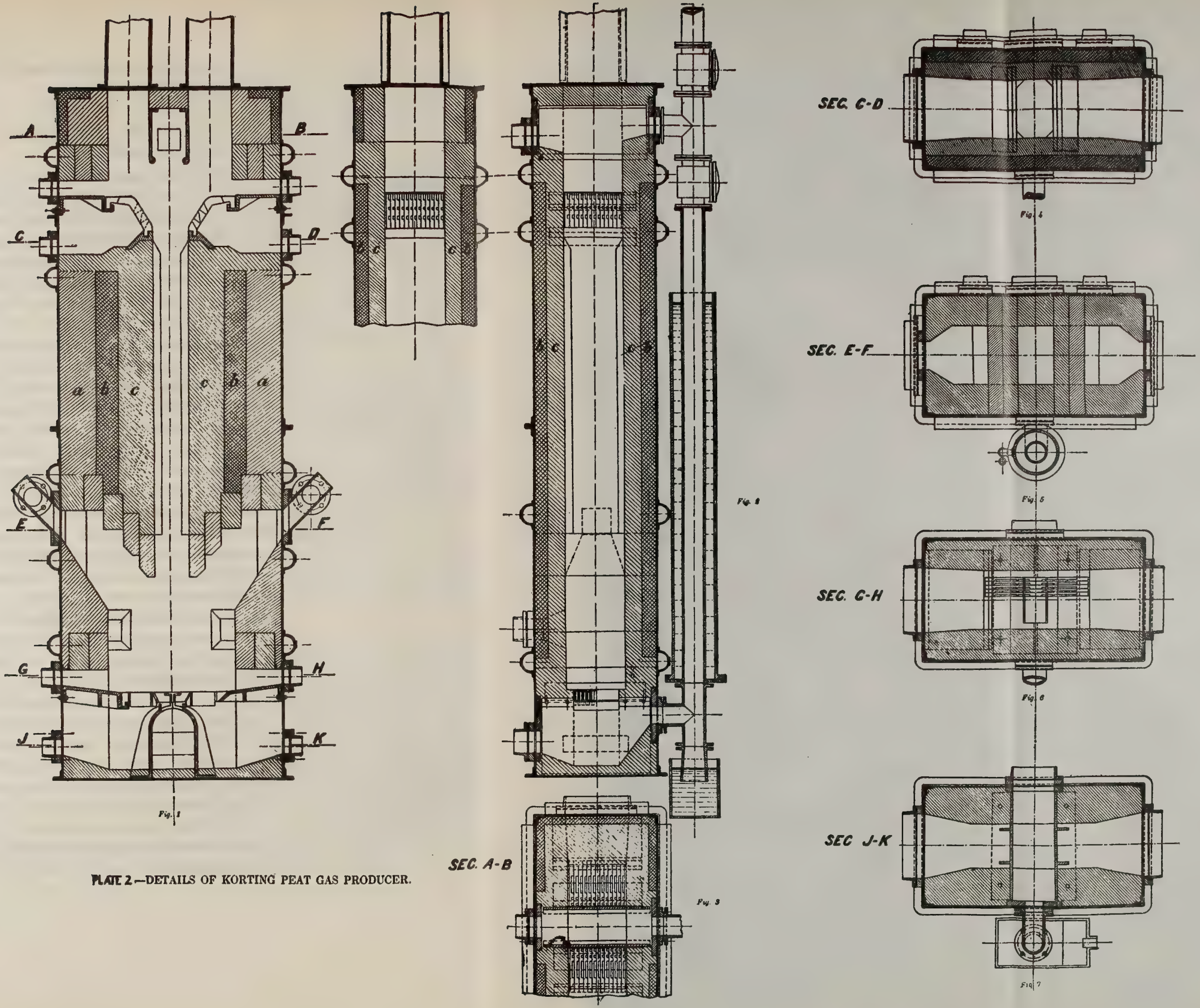
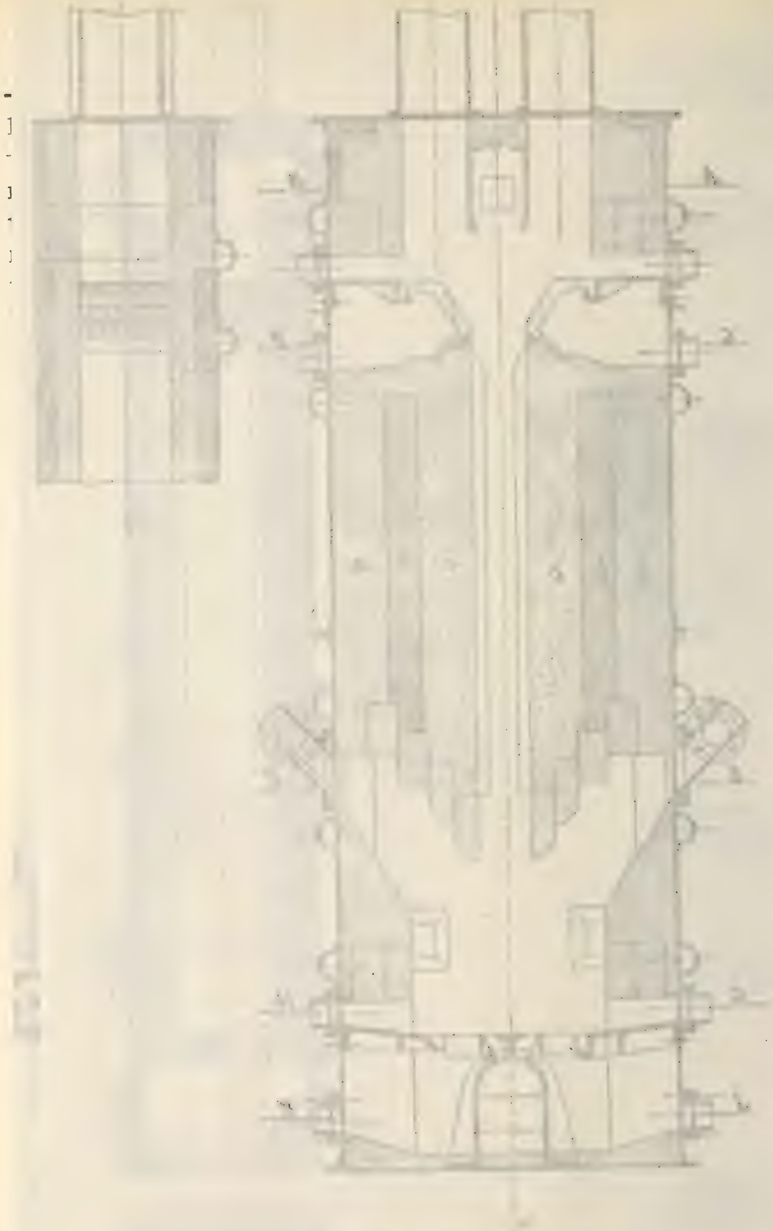


PLATE 2.—DETAILS OF KORTING PEAT GAS PRODUCER.



[illegible]

THE GAS-CLEANING SYSTEM OF THE PLANT PRESENTED THE FOLLOWING DISADVANTAGES

(1) The gas-cleaning system, while removing a considerable amount of matter from the gas, failed to remove the tar which caused the trouble at the engine. The gas carried past the entire system a considerable quantity of a liquid tar in the form of vapor, which condensed on the valves, cylinder and piston rings of the engine, causing them to stick.

(2) A considerable amount of light tarry matter of a yellow color was deposited in the off-takes M, Fig. 4, which after a few hours of running filled the orifice of this off-take to such an extent that the suction was very greatly increased.

(3) Some of the tarry matter adhering to the outside of the inside cylinder K-K, was washed off, and dropped to the bottom of this portion of the scrubber, between the enclosing and enclosed cylinders. When the gas was very dirty this accumulation was considerable, and had to be removed after a few days' operation.

(4) The baffle-plates of the tar filter became in a short time so choked with matter passing the wet scrubber, that the engine operated with difficulty on account of the high suction.

(5) While the increase in suction on the gas main due to these causes did not often necessitate the shutting down of the engine when the engine was once started — the greatest difficulty was experienced in starting on a cold system.

Under the circumstances above enumerated, the plant could hardly be called practical or commercial in any sense, since the amount of time and labor involved in cleaning the different parts of the system was excessive. To overcome these difficulties and, if possible, render the plant entirely commercial and practical in every respect, considerable time was devoted to investigation of the nature of the tarry products carried by the gas leaving the producer, and experimentation with different devices for removing them. As a result of this work, the writer devised a modification to the wet coke scrubber and tar filter which proved successful in removing the deleterious matter from the gas — and this economy was achieved without increasing the suction on the engine.

MODIFICATION TO WET COKE SCRUBBER AND TAR FILTER

The modification devised by the writer to the wet coke scrubber is shown in Fig. 6 — and consists of a fine mesh screen cone which takes the place of the perforated cylinder previously described. The cold water spray L, now washes off the matter deposited on the outside of the cone to the bottom, whence it leaves by the overflow provided for this purpose. Inside the cone is placed an additional spray provided with hot and cold water connections. In practice it has not been found necessary to use the hot water on this spray — it was provided for the purpose of reducing excessive suctions which might arise through the clogging of the fine meshes of the screen.

The tar filter was altered by removing the baffle plates and four perforated plates, and introducing in their place, four screens varying from coarse mesh at the gas inlet end to very fine mesh at the gas outlet end. These screens were so constructed that they could be easily taken out by removing the cover-plate of the filter. A spray, provided with both hot and cold water connections, is placed over each of these screens. This method of cleaning the screens has proved to be very efficient and the hot water connection can be dispensed with, while running.

ADVANTAGES OF IMPROVED CLEANING SYSTEM

The improved cleaning system possesses the following advantages:

(1) The wet coke scrubber completely clears itself of any matter, thus reducing the suction at this point to a minimum, and retaining it throughout a run of a period of considerable duration.

(2) The larger portion of the heavier tarry matter is separated from the gas in this scrubber.

(3) The tar filter separates out the more liquid or lighter tars, and does not clog. The suction at this point is very low, and does not vary to any extent throughout a run of almost any duration.

(4) By means of these modifications the total suction on the gas main leading to the engine has been reduced from 6 or 7 inches, and more to about 3 inches, with the resulting greater efficiency in cleaning the gas.

As the plant is at present constructed and operated, very little tarry matter is carried past the producer, hence but comparatively little material, possessing high heating value, is lost in the cleaning system.

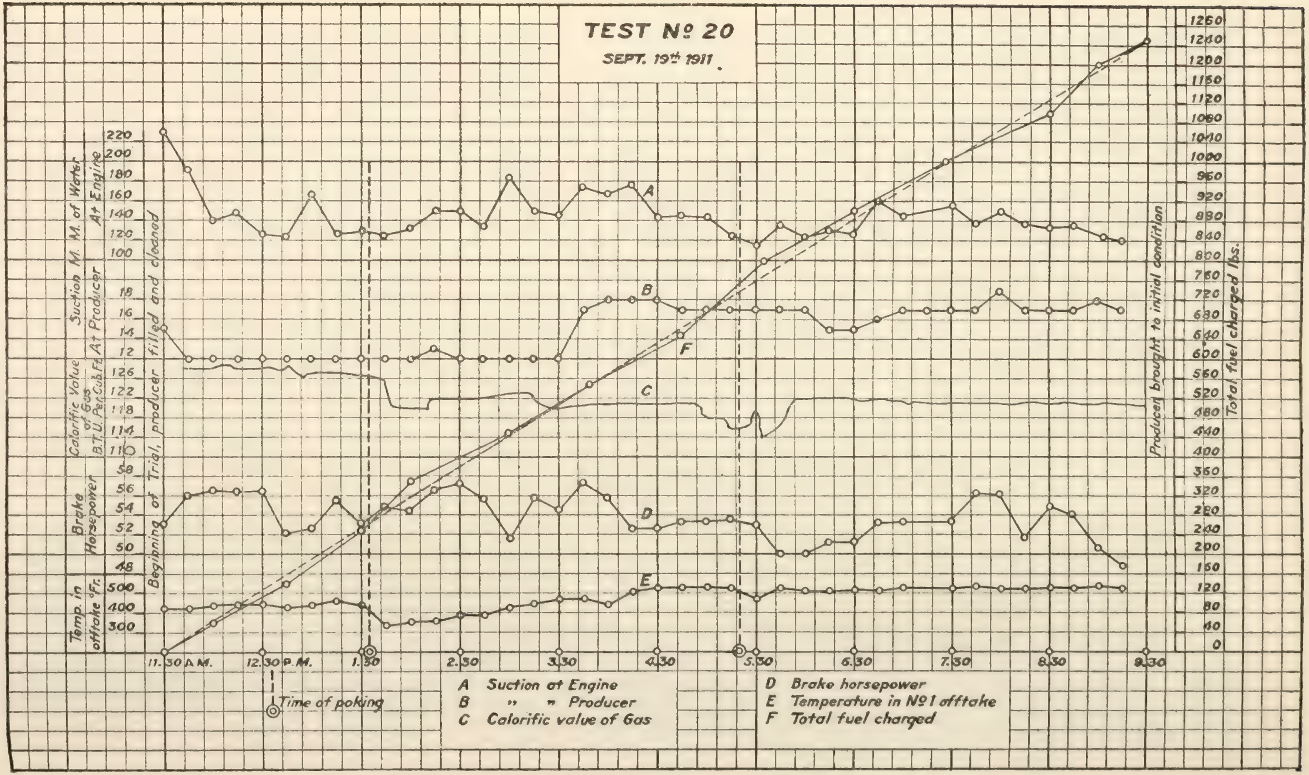
The engine can be operated at any load up to maximum for a long period, either continuously or intermittently, without the necessity of cleaning valves or piston. Previously, it was found necessary to wash the piston several times a day with a mixture of oil-soap and water; and although this method of cleaning the cylinder and piston of tarry matter proved reliable and efficient, the operation was always more or less dirty. The tar distilled from peat at the temperatures prevailing in the producer is soluble in a mixture of oil-soap and water; and since no harm results from employing this method of cleaning while the engine is in operation, the plant, strictly speaking, was commercial, even under these adverse conditions.

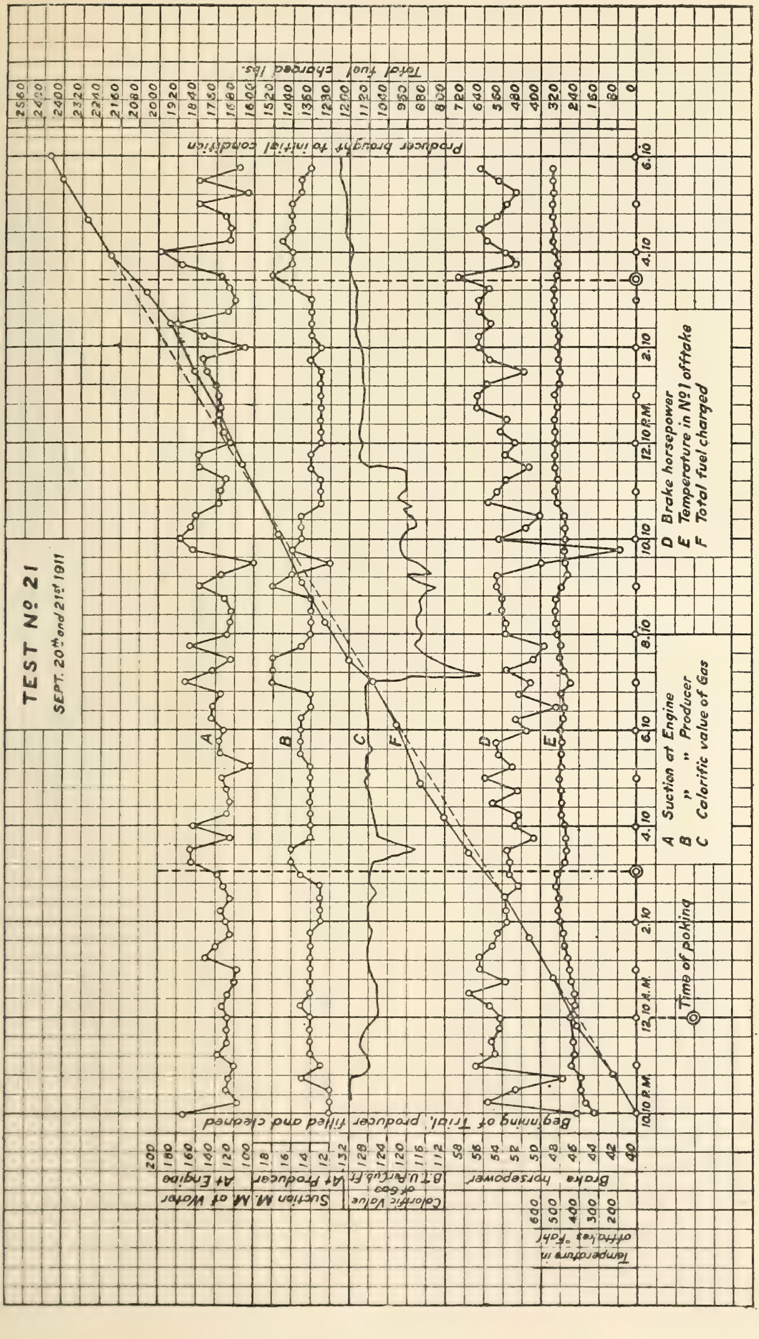
RESULTS OF TESTS

The following summary of the results of a 20 and 10-hour test at a load of about 53 B. H. P. made with the 60 B. H. P. Körting producer-gas power plant at the Fuel Testing Station, Ottawa, and the Chart No. 1, which shows the results of the 20-hour test graphically are self-explanatory and require but few comments.

TRIALS WITH ALFRED PEAT, AIR OPENING FOR FULL LOAD ON GAUGE

1. No. of trial,	20		
2. Date of trial,	Sept. 19	Sept. 20 and 21	1911
3. Time of starting,	11.30 A.M.	10.10 A.M.	
4. Time of stopping,	9.30 P.M.	6.10 A.M.	
5. Duration,	10	20	hrs.
6. Total peat charged during trial,	1250	2450	lbs.
7. Total ash and clinker drawn during trial,	60	116	lbs.
8. Total peat used for banking and starting,		431	lbs.
9. Total peat used during trial and for banking and starting,		2881	lbs.


TEST No. 20
SEPT. 19th 1911.
Suction M. M. of Water At Engine
220
200
180
160
140
120
100
At Producer
18
16
14
12
Calorific Value of Gas B.T.U. Per Cub. Ft.
126
122
118
114
110
Brake Horsepower
58
56
54
52
50
48
Temp. in offtake °Fr.
500
400
300
Beginning of Trial, producer filled and cleaned
Producer brought to initial condition
Total fuel charged lbs.
1260
1240
1200
1160
1120
1080
1040
1000
960
920
880
840
800
760
720
680
640
600
560
520
480
440
400
360
320
280
240
200
160
120
80
40
0
11.30 A.M.
12.30 P.M.
1.30
2.30
3.30
4.30
5.30
6.30
7.30
8.30
9.30
Time of poking
A
B
C
D
E
F
A Suction at Engine
B " " Producer
C Calorific value of Gas
D Brake horsepower
E Temperature in No. 1 offtake
F Total fuel charged


TEST No. 21
SEPT. 20th and 21st 1911
A Suction at Engine
B " " Producer
C Calorific value of Gas
D Brake horsepower
E Temperature in No. 1 offtake
F Total fuel charged
Time of poking
Beginning of Trial, producer filled and cleaned
Producer brought to initial condition
Total fuel charged lbs.
Suction M.M. of Water At Engine At Producer
Calorific Value of Gas B.T.U. Per Cub. Ft.
Brake horsepower
Temperature in offtakes °Fahr.
10.10 P.M.
12.10 A.M.
2.10
4.10
6.10
8.10
10.10
12.10 P.M.
2.10
4.10
6.10
A
B
C
D
E
F
200
180
160
140
120
100
18
16
14
12
132
128
124
120
116
112
58
56
54
52
50
48
46
44
42
40
600
500
400
300
200
2560
2480
2400
2320
2240
2160
2080
2000
1920
1840
1760
1680
1600
1520
1440
1360
1280
1200
1120
1040
960
880
800
720
640
560
480
400
320
240
160
80
0

Particulars of Peat Used

10. Moisture % in peat as charged,	31.4	30.2	

Proximate Analysis of Dried Peat as Charged

11. Fixed carbon,	29.5	30.0	%
12. Volatile matter,	64.8	64.6	%
13. Ash,	5.7	5.4	%
14. Calorific value of dry peat,	9470	9450	%
14. Calorific value of dry peat,	9470	9450	B.T.U.
15. Calorific value of peat as charged,	6500	6600	per lb.
16. Combustible matter in refuse,	53.4	29.7	%
17. Barometer reading,	29.85	29.88	inches
18. Wet bulb (in producer room),		59.2	F
19. Dry bulb,		67.1	F
20. Humidity,		64.8	%
21. Average suction at producer,	.6	.5	in. of
22. Average suction after coke scrubber,	4.3	2.6	water
23. Average suction after tar filter,	4.7	4.7	in. of water
24. Average suction after dry scrubber,	5.7	5.4	in. of water
25. Temperature of gas leaving producer in No. 1 exit	469	475	F.

Engine and Generator

26. Average revolutions per minute of engine	190	190	
27. Temperature of outlet cooling water,	111	115	F.
28. Average kilowatts delivered to switchboard,	35.3	35.2	
29. Average electrical horsepower,	47.3	47.2	
30. Efficiency of dynamo,	.88	.88	
31. Average brake horsepower of engine,	53.8	53.7	

Gas Analysis Per Cent by Volume

	10 samples	20 samples	
Sample taken after passing tar filter.			
32. Carbon dioxide,	10.8	9.9	%
33. Ethylene,	.5	.4	%
34. Oxygen,	.5	.3	%
35. Carbon monoxide,	17.7	20.2	%
36. Methane,	2.4	2.4	%
37. Hydrogen,	10.2	10.3	%
38. Nitrogen,	57.9	56.5	%
39. Inflammable gas,	30.8	33.3	%
40. Calorific value from analysis (Gross),	122	128	B.T.U. per cu. ft.
41. Calorific value from analysis (Net),	113	120	
42. Average net calorific value from recording gas calorimeter,	123	127	
Results of Tests			
43. Total peat charged during trial,	1250	2450	lbs.
44. Total dry peat charged during trial,	858	1710	lbs.
45. Total ash and clinker drawn during trial,	60	116	lbs.
46. Ash and clinker drawn per cent of peat charged,	4.8	4.7	
47. Ash and clinker drawn per cent of dry peat charged,	7.0	6.8	
48. Average kilowatts delivered to switchboard,	35.3	35.2	
49. Average brake horsepower of engine,	53.8	53.7	
Hourly Quantities			
50. Lbs. of peat charged	125	122	
51. Lbs. of dry peat charged,	86	86	

Economic Results			
52. Peat charged per K. W. hour,	3.54	3.47	lbs.
53. Dry peat charged per K. W. hour,	2.44	2.44	lbs.
54. Peat charged per B. H. P. hour,	2.32	2.27	lbs.
55. Dry peat charged per B. H. P. hour,	1.6	1.6	lbs.
56. Overall thermal efficiency of engine and producer,	16.8	16.9	%
Results Deduced from Fuel and Gas Analyses			
57. Air supplied to producer per lb. of dried peat charged,	39.2	37.6	cu. ft.
58. Water supplied to producer per lb. of dried peat charged,	.47	.46	lbs.
59. Cubic ft. of gas produced per lb. of dried peat charged,	53.4	52.6	
60. Heat equivalent of gas produced per lb. of dried peat charged,	6040	6310	B.T.U.
61. Producer efficiency,	63.8	66.8	%
62. Cubic ft. of gas delivered to engine per hour,	4590	4520	
63. Cubic ft. of gas delivered per B. H. P. per hour,	85	84	
64. Heat equivalent of gas delivered per B.H.P. per hour,	9660	10100	B.T.U.
65. Thermal efficiency of engine (B. H. P. basis),	26.3	25.2	%

The gas-cleaning system used during these tests was not yet perfected and although the gas delivered to the engine was sufficiently clean, the fluctuations in suction at the wet coke scrubber were quite noticeable. The tar filter, moreover, had not yet been altered and some considerable fluctuation was observed due to clogging of this portion of the cleaning system.

Since altering both the wet coke scrubber and tar filter a noticeable reduction in the total suction has been observed. This suction does not vary more than a few centimeters during a 10-hour run and no trouble is experienced when starting up on a cold system. Since the tests above quoted were made, the screen cone in the wet coke scrubber has been replaced by one of coarser mesh which in conjunction with the screens in the tar filter has proved entirely efficient and satisfactory both as regards the resistance set up in the system due to these screens and the cleanliness of the gas.

The total suction on the engine main varies but little from three inches and often falls below this amount.

Quality of the Gas: The chemical composition and calorific value of the gas is only slightly altered as a result of cleaning the fires. When one side is poked at a time this variation is reduced to a minimum which is not of sufficient amount to necessitate resetting of the air and gas valves of the engine.

Operation of the Producer: The operation of the producer as it is now designed and with the grates of larger openings over the gas chamber K. Fig. 1, substituted for those of newer design, is very uniform. The feeding of the fuel from the upper zone proceeds regularly and evenly and the peat fed to the lower zone appears to be thoroughly coked. The producer can be operated for several hours without poking or cleaning the fires and since this operation as well as that of removing the ashes can be performed while the plant is in operation, the producer is capable, in every respect, of continuous operation.

Cleaning of the Producer Gas Mains, etc.: The gas pipe leading the gases evolved at the upper zone to the lower zone should be cleaned once a week — owing to the accumulation of tar in the damper chambers while the producer is standing idle. The accumulation, mostly pitch, in the two gas off-takes is of small amount during a week's run — but to insure the proper operation of the producer these pipes should be cleaned about once a month.

Reliability: The peat producer-gas power plant as now constructed may be pronounced as thoroughly reliable. Its operation may be carried on continuously for a week or more without having to shut down for the purpose of cleaning the valves of the engine. The engine has been operated for a period of 150 hours without removing either the admission or mixing valves for cleaning, and it should not be found necessary to remove the piston, in commercial practice, more than once in six months for purposes of cleaning.

Attendance: A peat producer-gas power plant of the capacity installed in the Fuel Testing Station can safely be left in the hands of an intelligent laborer after he has received instructions in the handling of the plant for a week or so, from a competent engineer. The services of only one man is required to run such a plant when it is operated on day shift work only.

Fuel Consumption: The consumption of fuel per Brake Horsepower hour, including stand-by losses, is for full load 1.7 lbs. of dry peat, or 2.3 lbs. of peat containing 25% moisture; for 3/4 load the fuel consumption including stand-by losses is 2.1 lbs. of dry peat, or 2.8 lbs. of peat containing 25% moisture. The fuel consumed while the producer stands idle has been determined for a period extending over several months. The average consumption has been found to be between 2 and 3 lbs. per hour when the producer stands over night, *i.e.*, for 14 hours; and between 1 and 2 lbs. per hour for longer periods, *e.g.*, three days or more.

Cooling Water: The amount of water used for cooling and cleaning the gas is normal.

COST OF FUEL

In estimating fuel costs, the assumption is made that peat with a moisture content of 25% can be delivered to the producer for $2.00 per ton. In order, however, to take advantage of this, or a lower cost for fuel, the power plant must be situated at or near the bog where the peat fuel is manufactured. For small plants of the type and capacity described, this might not prove feasible in many cases — but will prove entirely feasible and practicable when the plants erected are of large capacity, and when the energy generated is transmitted in the form of electricity to neighboring towns and villages, for lighting, power, and other purposes.

Since the fuel burned in the producer does not require to be of the best quality, the fuel cost may be considerably reduced; since the broken peat bricks, and considerable fines, which always occur in the manufacture of peat, and which otherwise would represent a loss, can be efficiently utilized in the producer. Assuming, however, that peat can be delivered to the plant for $2.00 per ton, and that the plant is running with a power factor of 75% for 3000 hours, the fuel costs would be $8.40 per B.H.P. year, including stand-by losses.

ERRORS IN THE DETERMINATION OF MOISTURE IN COAL

By W. F. Hillebrand and W. L. Badger
Bureau of Standards, Washington, D. C.

The senior author was asked by the President of a sub-committee[1] of the International Committee on Analyses to co-operate in the determination of moisture in six samples of coal.

A large number of determinations of moisture were made on these samples, but the work was finished too late to be incorporated in the committee's report. A comparison of the results obtained by different methods showed the influence of the various factors which tend to affect the results. The magnitude of these effects is not generally recognized, in spite of the considerable amount of work that has been done in recent years. It seems therefore worth while to present here a typical series of the results obtained and to discuss briefly the subject of moisture determinations in coals, a subject which has interested one of us for many years and has been extensively studied by those connected with the fuel testing work of the United States Government at and subsequent to the Louisiana Purchase Exposition at St. Louis, in 1904, and to a less extent by others.

Methods for determining moisture, in general, belong to the class of indirect methods, which in the absence of a simple reagent for water are most convenient and expeditious; but when applied to so complex and easily oxidized a substance as coal, a number of reactions occur which greatly modify the results. In general these reactions are:

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[1] On the Standardization of Methods of Determining Water in Coal and other Fuels and in Minerals.

As described in the letter of the President of the sub-committee, Mr. George T. Holloway, these coals are:

No. 1. Anthracite from South Wales.

No. 2. From the "10-yard" or "thick coal" seam of South Staffordshire, which in many respects resembles a lignite.

No. 3. From the Kilburn seam in Leicestershire (England). A coking coal largely used for general and household purposes.

No. 4. From South America, interesting in connection with moisture determination on account of its high sulphur content (4.2% volatile and 8.2% non-volatile). No pyrites is visible or can be washed out, but the coal rapidly weathers and develops free sulphuric acid and ferrous sulphate.

No. 5. An ordinary bituminous coal from Leicestershire.

No. 6. A somewhat less bituminous coal from the southeast of Scotland.

(*a*) The sensitiveness of coal to atmospheric conditions, especially when finely powdered.

(*b*) The giving off of volatile substances other than water.

(*c*) The absorption of oxygen, which may take the form of:

1. Oxygen added directly to the coal substance.
2. Oxygen combined with carbon and split off as CO_2.
3. Oxygen combined with hydrogen and split off as H_2O.

All of these reactions have been recognized by many investigators, and though they may not appear with equal prominence in all coals, undoubtedly most or all of them take place simultaneously in all cases, with one or another predominating.

It is now customary in this country to prepare coal samples in two stages: first, coarse crushing followed by air drying, and second, fine grinding of the air-dried sample. It is generally recognized that the distinction between the "air-drying loss" and the moisture as determined on the air-dried sample is not a definite one. Fineness, area of surface exposed, temperature, and especially the humidity of the air, all affect the amount of loss at this stage. It is perhaps not so generally recognized that, if the test is prolonged or if carried out on finely ground coal, considerable oxidation may take place. This was noted in a number of experiments made in the laboratory of the U. S. Geological Survey at St. Louis.[1] Our experiments on the British and South American Coals bear out this point. These had been air-dried in England, but proved to contain more moisture than was in equilibrium with the air of the laboratory in Washington, as shown by the following air-drying tests. The first series was made in glass weighing bottles, 4 x 6 cm., containing about 25 grams coal (half full); the other on samples of 9–10 grams' weight spread out on 9-cm. watch glasses. They were weighed at intervals until they were practically in equilibrium with the air of the laboratory, with the following results:

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[1] Bull. U. S. Geol. Survey, No. 232, pp. 13-16.

Coal No.	I	II	III	IV	V	VI
Loss in bottles	1.52	5.17	4.18	1.60	3.50	4.82
Loss on watch glasses	2.38	7.78	5.55	2.94	5.52	6.69

These results illustrate in a striking manner the influence of the atmospheric humidity. The air-drying was done in Washington in winter, when the humidity was low, ranging approximately from 15 to 25 per cent. When the results are compared with the eight-day tests over sulphuric acid (p. 7), it is seen that the losses represent in a number of cases over 50 per cent of the moisture that was present when the coals were bottled in England, even in the series where least oxidation had taken place during air-drying (the first of the above and II of p. 7).

In the moisture determinations made on these two sets of air-dry samples, the second set gave higher results than the first in every instance; but the difference was only a few tenths of one per cent instead of the one or two per cent differences shown in the air-drying losses. This would seem to indicate that the second set, with its greater surface exposed, took up more oxygen than the first set, giving it off to a certain extent as H_2O and CO_2.

A series of moisture determinations was also made on the coals without air-drying (see III of p. 7). This set of results was with some coals still higher than on either of the previous sets; indicating that there was some gain in weight of the coals due to oxidation in both of those. In other words, every exposure of finely ground coal to the air is not only accompanied (a) by a loss of water, if humidity conditions are such that the moisture in the coal is not in equilibrium with the moisture in the air, but also (b) by an absorption of oxygen, which may take the form of a gain or loss in weight, according to whether fixing of oxygen by the unsaturated coal substance or loss of hydrogen and carbon as water and carbon dioxide is a predominating reaction.

The second factor — loss of volatile matter other than water — depends largely on the fineness of the coal and the length of time that has elapsed since it was mined. Even coarsely crushed coals give off large amounts of methane and traces of other volatile substances, but this loss takes place, in great part, immediately after the coal has been mined. It would seem that the loss in weight so caused, during the ordinary process of air-drying previous to analysis, would be comparatively slight; both because most coals are sampled sufficiently long after mining to allow such losses to take place before the sample was taken, and also because the air-drying is generally carried out on a sample that has been already partially crushed. That there is a loss of gases on grinding the air-dried sample is a point that is yet to be determined. In the same way, though freshly mined coals absorb oxygen with avidity even when coarse, after the time which generally elapses between mining and sampling it would seem probable that the velocity of this reaction would have fallen off to the point where the coarse sample would absorb comparatively small amounts of oxygen during air-drying. Prolonged exposure to the air, or exposure of a finely ground sample, is, however, by no means free from liability to serious error.

The methods used for the analysis of this final air-dried sample may be divided into vacuum methods and oven methods. In our work we used:

1. Drying in a vacuum over sulphuric acid for 1, 2 and 8 days.
2. Drying for one hour in an oven at 105-110°.
3. Drying for two hours in an oven at 105-110°.
4. Drying for one hour at 105-110° in a current of dry air.

The six coals were of very different types, but the effect of conditions was the same within very narrow limits on all of them, no matter what the method of previous preparation had been, and the results are in entire agreement with the senior author's past experience. In the vacuum method two of the factors — equilibrium with the surrounding air, and oxidation — are removed, and the only disturbing condition is the loss of volatile matter other than water. Owing to the fact that no darkening of the sulphuric acid takes place it is probable that little of the higher hydrocarbons is given off; and the fact that a vacuum may be maintained without change indicates that little methane is evolved. The possibility of volatile material being evolved during evacuation of the desiccator is not precluded. This method gave the highest figures, which are probably nearest the truth, though a slow loss of moisture continued with some coals up to the eighth day and might have continued longer. Exposure for 24 hours was in no case sufficient.

In drying in the oven all three disturbing factors come into play. The air in the oven becomes saturated with moisture, so that an equilibrium is reached before the coal is perfectly dry, and this point varies from day to day with the varying humidity. An opportunity for oxidation is presented, and though part of this oxidation certainly goes to form volatile products (CO_2 and H_2O) the amount of oxygen absorbed as such is sufficient to cause a net gain in weight of the dry coal substance. This is shown by the fact that in all cases the figure obtained by this method was the lowest of all. The error caused by oxidation is sufficient to mask any effects due to volatilization of light hydrocarbons, though such a distillation certainly occurs. On prolonging the heating for two hours, little change takes place; a net gain of loss of a few hundredths of a per cent was noticed, in one direction with some coals and opposite with others.

The modification of this method by the introduction of a stream of dry air gives much more consistent figures and much higher ones, approaching very nearly the results obtained in the vacuum. This is obviously due to the fact that the water is removed as fast as liberated, thus preventing the coal from attaining an equilibrium with the air of the oven before all its moisture has been removed. The greater uniformity so obtained is best shown by a few figures. A sub-bituminous coal gave, as the average of 7 determinations without dry air, 9.46%, with a maximum variation of 0.83%. With a current of dry air, the average of 8 determinations was 9.93%, with a maximum variation of 0.23%. Similar results were obtained on all types of coal.[1]

In all, about 300 individual determinations were made on the six coals, and the results were remarkably uniform in their indication of the effects of the various disturbing factors outlined above. It will be of interest to compare the figures for these coals to illustrate the range of values possible. Series II was air-dried in glass weighing bottles, Series III on watch glasses (as discussed on

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[1] Archibald and Lawrence's tests (*J. Ind. Eng. Chem.*, *4*, 258, 1912) support with quantitative data, the well-established fact that the usual official method fails to give, as a rule, anything like the true moisture content of coals; but their recommendation to substitute for it drying in a current of dry air and direct weighing of the water is open to criticism as an exact method, for the reason that marked oxidation of the coal takes place and water is undoubtedly produced as a result.

p. 3), and Series IV was taken direct from the containers in which the coal was sent us. This last series represents far more nearly the moisture condition of samples as received.[1]

RESULTS OF MOISTURE DETERMINATION ON FIVE BRITISH AND ONE SOUTH AMERICAN COAL

Coal	Series	Air-drying loss	Vacuum over H_2SO_4			Oven without dry air		Oven dry air
			1 day	2 days	8 days	1 hr.	2 hrs.	1 hr.
I	II	1.52	2.33	2.36	2.38	2.30	2.34	2.41
	III	2.38	2.88	2.89	2.90	3.08		
	IV				2.50	2.40	2.47	2.40
II	II	5.17	9.43	9.69	9.90	9.38	9.47	9.78
	III	7.78	9.98	10.16	10.33	10.45		
	IV				10.14	9.54	9.68	9.93
III	II	4.18	6.97	7.31	7.38	6.96	7.02	7.32
	III	5.55	7.45	7.62	7.76	7.51		
	IV				7.28	6.44	6.77	6.93
IV	II	1.60	4.77	5.10	5.56	5.24	5.54	5.72
	III	2.94	5.26	5.50	6.07	5.62		
	IV				5.86	5.17	5.49	5.88
V	II	3.50	7.09	7.31	7.46	7.04	7.17	7.38
	III	5.52	7.52	7.68	7.80	7.56		
	IV				7.30	6.48	6.56	6.80
VI	II	4.82	7.30	7.48	7.58	7.18	7.21	7.43
	III	6.29	7.82	7.94	7.99	7.62		
	IV				7.46	6.69	6.75	6.89

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[1] Since this paper was written a copy of the report of the sub-committee referred to has been received from Mr. Holloway. Our attention is attracted by the disagreement of our own results for Coal No. 1, Series III excepted (by all of our methods), with those of the other analysts, all of whom found much more moisture in this coal. Our sample, when it reached us, had most certainly a much lower moisture content than the others. The data of Series III as to the vacuum and 1-hour tests seem to contradict this statement, but, as said above, the samples of this series had undoubtedly suffered much oxidation during their exposure for air-drying.

In view of the wide discrepancies between the different indirect methods, a direct one is very desirable. Work has been done by us on such a method, but as yet no definite conclusions have been reached. In the first attempts the coal was heated in a U-tube in a current of dry hydrogen and the moisture was absorbed in sulphuric acid. A number of results were obtained, but later investigation showed the presence of some errors which render their value questionable. It may be said in general, however, that the figures so obtained were higher than by any of the other methods. In every case the loss in weight of the coal was slightly less than the gain in weight of the acid. This may be ascribed to the presence of oxygen in the hydrogen, or to absorption of hydrogen by the coal substance, or to a splitting off of hydroxyl from the coal to form water with the hydrogen. Later, work was begun on an apparatus for heating the sample in a stream of pure nitrogen, which is still under way. Considerable difficulty has been experienced in obtaining perfectly satisfactory blanks, but it is hoped that when perfected the method will permit of a direct and absolute determination of water. In some preliminary experiments the coal, even in nitrogen, showed less loss in weight than corresponded to the amount of water obtained, as was earlier observed by Porter and Ovitz.[1] No explanation is offered at present for this behavior.

Other direct methods which might prove of value are some modification of the carbide method, or of the magnesium-methyl-iodide method, or heating in a high vacuum, with direct weighing of the water. All these methods have been considered at the Bureau of Standards and it is hoped that opportunity will be found to make an exhaustive comparative study of them, with the object of finding some direct primary method which, even if too involved for general use, may yet serve as a standard of reference.

Our work thus far has been directed toward fixing a method for the exact determination of moisture in the laboratory sample. The securing of a laboratory sample that shall, in connection with the preliminary air-drying of the coarsely crushed sample, give the true water content of the coal, is another problem, one on which the authors and others are engaged in connection with the work of the Coal Committee of the American Chemical Society.

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[1] *J. Am. Chem. Soc.*, *30*, 1486 (1908). This observation is also in agreement with that of Archibald and Lawrence (*J. Ind. Eng. Chem.*, *4*, 258, 1912) on a bituminous coal at 110° and 120°; but not at 75° and 100°, nor at any one of the four temperatures on an anthracite, in all of which cases the loss in weight of coal exceeded the weight of the moisture weighed.

A NEW COKING TEST FOR FUELS

By R. Lessing, Ph.D., F.C.S.
London, England

At the Seventh International Congress of Applied Chemistry held in London in 1909 a paper was read by Prof. E. J. Constam on the estimation of volatile matter in fuels in which he gave a *résumé* of his experiences with various methods of carrying out the coking test. He came to the conclusion that the method of the American Committee on coal analysis (*Jour. Amer. Chem. Soc., 21,* 1122) gave better and more consistent results than the other methods under review and the proposal to accept this method as the standard for the yield of coke was carried and adopted as a resolution of the Congress.

The principal advantage of this method over others formerly in use is that it carries the heating far enough to allow the employment of temperatures high enough to ensure the complete carbonization of, and expulsion of volatile matter from, coal and other fuels.

There are, however, a number of drawbacks which still attach to this method and although they are known and recognised by workers in this field, their elimination has not, in spite of occasional attempts, hitherto been brought about.

The sources of error in the crucible test may be classed in two groups:

1. Factors tending to increase the "volatile" value and decrease the coke value. (*a*) Burning of coke in the more or less oxidising atmosphere. (*b*) "Spitting" of coal-dust due to explosive character of gasification of some coals or rush of currents produced by the flame.

2. Factors tending to decrease the "volatile" and increase the coke value. (*a*) Insufficient coking of the coal. (*b*) Secondary catalytic decomposition of the volatile products on the walls of the crucible. (*c*) Decomposition of volatile products by radia-

tion in the waste space of the crucible. (*d*) Deposition of carbon during coking by too rapid primary decomposition of volatile coal substance.

It has been repeatedly pointed out by various workers that results are dependent upon the size, shape, thickness and brightness of walls of the crucible as well as the atmosphere prevailing in it, and various modifications have been proposed to eliminate one or the other of these irritating factors. It can be said, however, that standardization of the method has not been reached.

I venture to submit a new method which, if not perfect in all its details, should go far in the standardization of the coking test of fuels. The technique of the method has been described in a paper recently read before the London Section of the Society of Chemical Industry (*Jour. Soc. Chem. Ind.* 1912, *3*, 465) and the following particulars may be briefly recalled:

In order to do away with the drawbacks due to the waste space above the coal and the excessive amount of heated surface and contact with the gas and vapours in the crucible test, the finely ground sample is heated in as compact a space as possible and out of contact with the air. For this purpose I used a cylindrical flat-bottomed tube made of quartz glass or platinum provided with a small flange on top by means of which it can be suspended. I have found an inner diameter of about 10 mm. to be a convenient size. One gram of powdered coal is weighed into this tube and a piston made of quartz glass tube or rod and loosely fitting into the outer tube is placed on top of the coal sample. By this means there is no waste space left above the coal and the gases and vapours evolved upon heating are expelled at the greatest possible speed, and have no chance of becoming stagnant and subject to the influence of radiant or contact heat for any length of time. I have found it convenient to effect the heating by placing the test tube into a wider tube made of quartz glass round which an electric resistance coil of platinum wire is wound. This arrangement enables me to apply the heat at any point desired and prevent the superheating of the products of carbonization. For details as to wiring I must refer to the paper quoted above.

The great advantages of this method lie not only in the fact that it gives an opportunity to standardize very exactly the shape

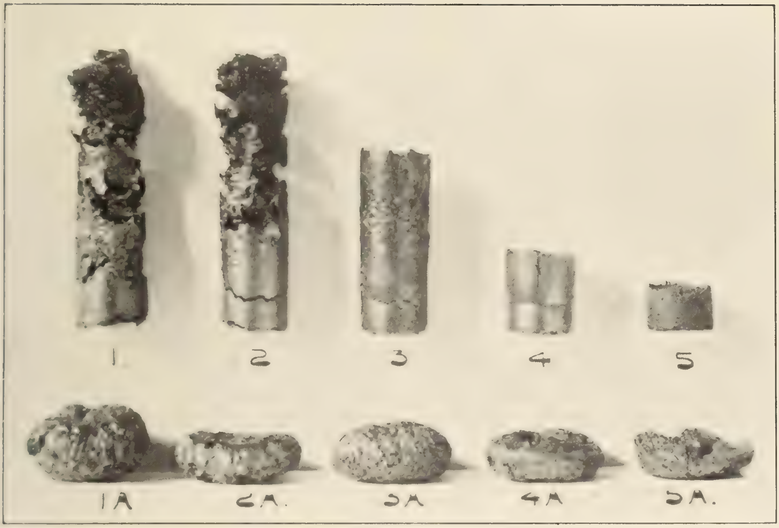


Figure 1.

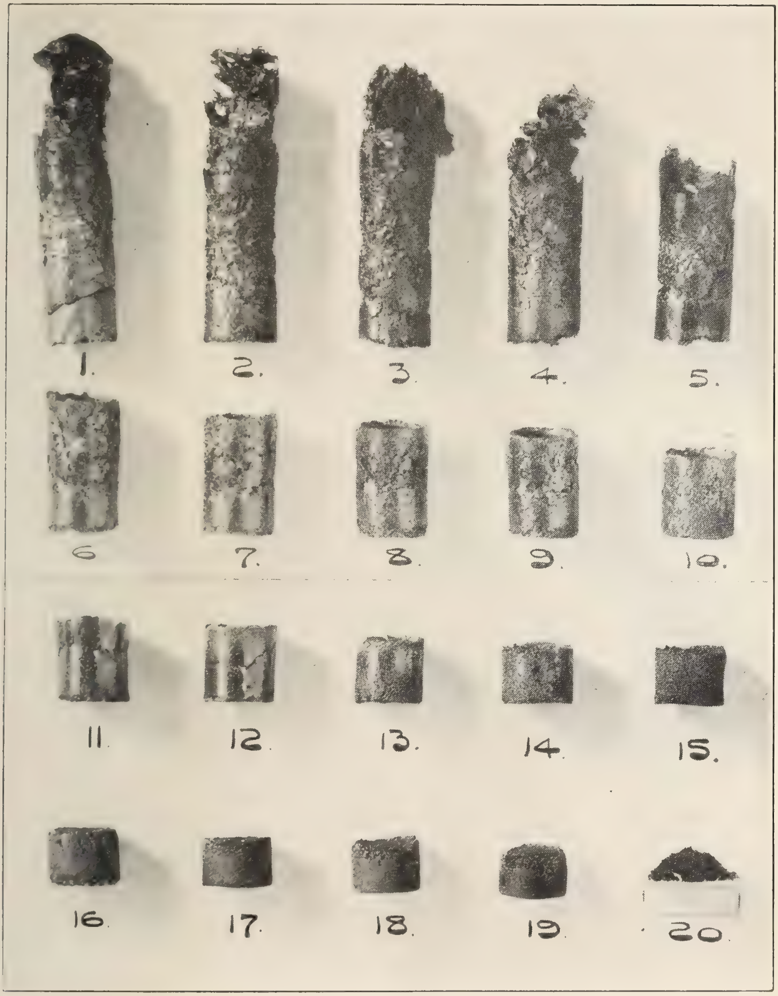


Figure 2.

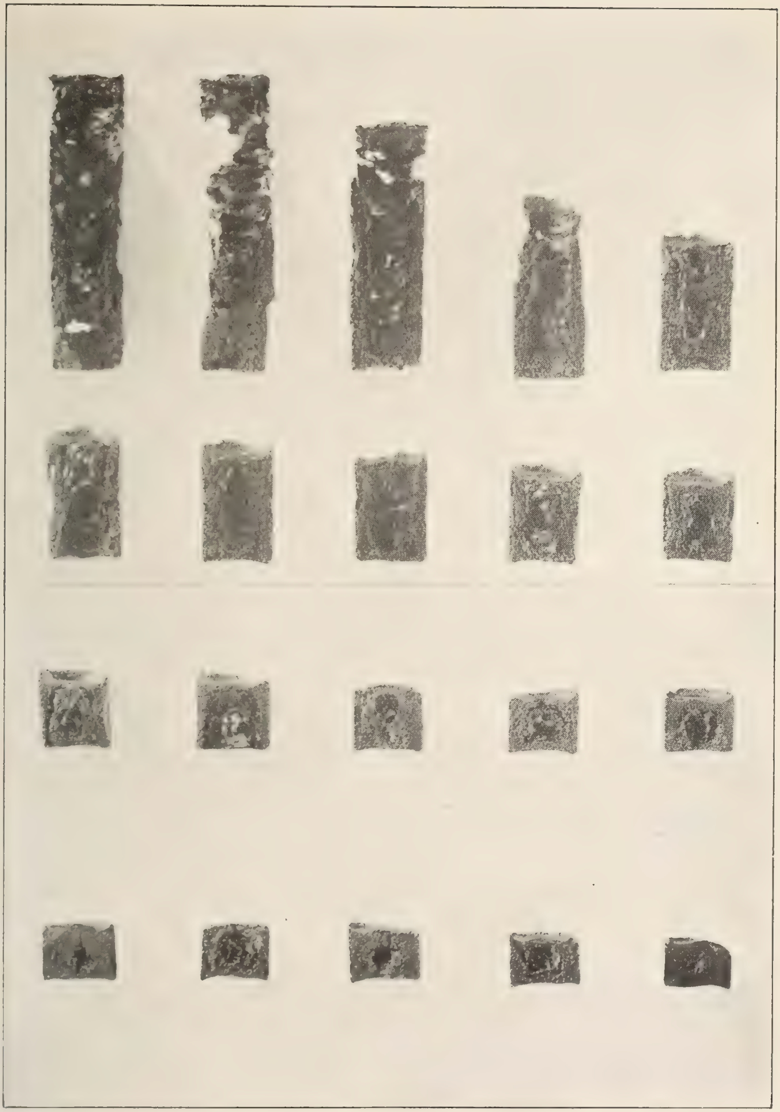


Figure 3.
A New Coking Test for Fuels.

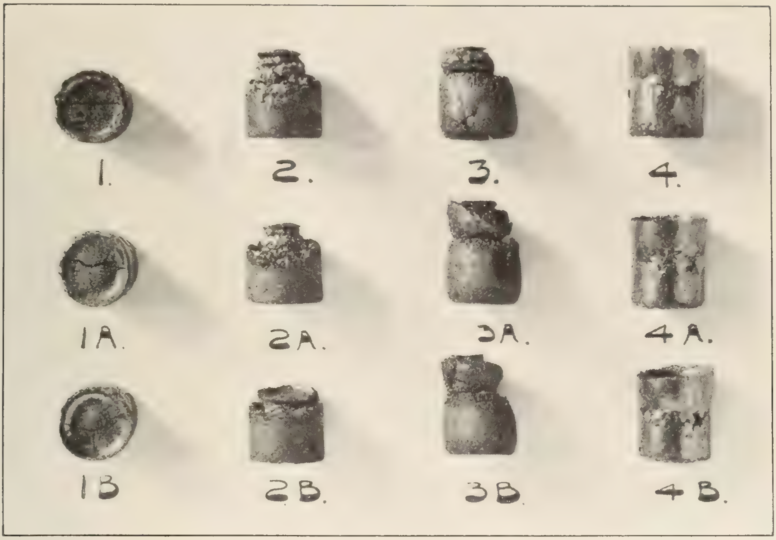


Figure 4.

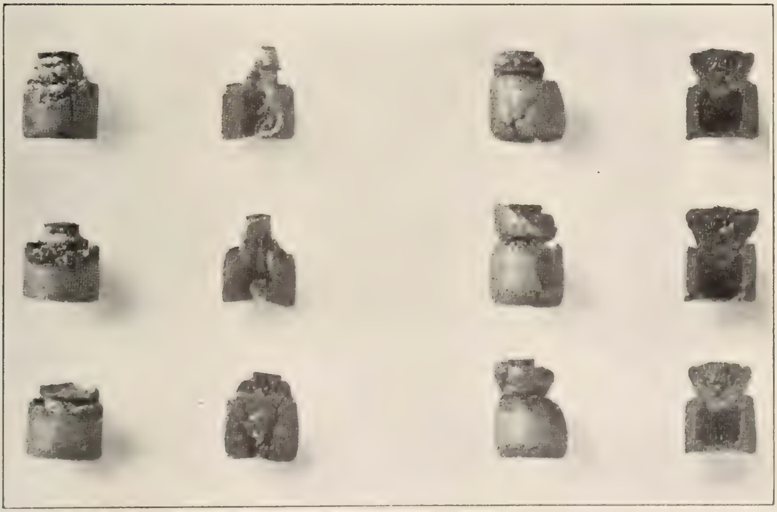


Figure 5.

and size of the reacting vessel and to do away with the waste space above the charge, but it also permits an insight into the coking process which could not be obtained with previous methods for the shape and aspect of the coke obtained is distinctive for every kind of coal.

Figure 1 shows the difference between the coke obtained in my new (top) and the old crucible method (bottom). That the differences between the cokes obtained from various coals are very marked will be seen still more clearly from Figure 2 in which photographs of the coke specimens obtained from twenty different coal samples are shown. One gram of the same fineness of each coal was employed and the proximate composition did not show very wide differences. It may be stated that even coal No. 20, which flatly refused to be coked, was not an anthracite but a bituminous coal containing about 32 per cent of volatile matter.

Figure 3 shows sections obtained from the coke specimens, and it is clear that the inner structure of the cokes will allow a still closer study of the nature of the coal samples and their behaviour upon carbonization.

From Figure 4 it will be seen that the results obtained by the method are not dependent upon chance, in which photos of three specimens each of the same sample are reproduced. It may be noticed that even in minute details of outer appearance specimens derived from the same coals are absolutely alike.

Figure 5 is still more striking in this respect as showing two sets of the samples of Figure 4 together with the corresponding sections.

The method can therefore practically be employed as one of identification of individual coals.

In a paper read before the Institution of Gas Engineers (see *Journal of Gas Lighting* 1912, *118*, 885) I have shown that the method can be carried further by closing the "retort" tube and collecting the volatile products. I have been able to ascertain that the yields of gas of certain coals correspond very closely with the yields known to have been obtained from the same coals on the large scale.

I venture to suggest that this method should be carefully and exhaustively tested by the workers in the province of fuel analysis and in view of the useful results obtained by the practical co-operation of members of the Sub-Committee for the determination of moisture in fuels and minerals of the International Committee on Analyses whose exhaustive report I have the honour to present to this Congress, I beg to propose that a similar committee be nominated for the comparative testing of methods for the estimation of volatile matter in fuels in order to bring about standardization and uniformity of testing.

I am aware that conditions for estimating volatile matter are still more complex than those attending the moisture determination but the results obtained in this case are encouraging enough to let us expect fruitful results, and I trust that my method will do something to further the efforts in this direction.

SUR LA COMBUSTION DES GAZ PAR INCANDESCENCE OU *COMBUSTION CONVERGENTE* ET SUR QUELQUES UNS DE SES EFFETS

PAR M. JEAN MEUNIER
Paris, France

La combustion par incandescence des gaz se distingue de leur combustion par flamme ou par explosion; elle nécessite pourtant aussi l'emploi des mélanges *explosifs* de gaz combustible et d'air. Le mélange qui alimente le bec Auer est formé d'air contenant 18 à 20% de gaz d'éclairage, proportion du maximum d'explosivité; si le gaz dépasse cette proportion, le manchon perd de son éclat et il n'éclaire presque plus.

Le simple examen de ces faits montre à l'évidence que le mélange combustible gazeux vient *converger* sur le corps solide incandescent et brûle en le frappant vivement. L'éclat est d'autant plus intense que l'énergie du choc est plus grande et le filament frappé plus fin; car ce filament est le foyer de la convergence, et, de même que pour les lentilles, plus le foyer est restreint plus le phénomène est brillant.

Il doit résulter de là les deux effets mécaniques moléculaires suivants:

1° La désagrégation subie à la longue par le corps solide porté à l'incandescence. Un manchon Auer ne peut durer qu'un certain nombre d'heures; sa désagrégation est manifeste.

2° Le choc doit produire le départ d'une partie du corps solide, c'est-à-dire sa sublimation.

Le platine, aussi bien le métal fondu que le métal aggloméré, qu'il soit pur ou allié au cuivre, possède la propriété d'entretenir la combustion convergente et de constituer la lampe sans flamme. Il suffit de le chauffer préalablement à 500 degrés environ. Pour un mélange de gaz et d'air à *teneur* déterminée, l'incandescence est d'autant plus *vive* que le fil de platine est plus fin. Celui-ci ne tarde pas à se désagréger et devenir gris, cassant et cristalin. Il est possible qu'il subisse une perte par sublimation, mais je n'ai pas encore essayé d'établir expérimentalement ce fait. En tout cas, le sulfate de chaux, déposé à sa surface en faible quantité, exalte en quelque sorte la propriété du platine et lui permet de se maintenir incandescent dans un mélange à 45% de gaz d'éclairage, tandis qu'autrement il devient obscur dans un mélange à 35%; mais ce sulfate de chaux disparait assez rapidement, sans manifestation apparente et sans que la température soit très élevée.

Le cuivre pur acquiert une propriété semblable à celle du platine, après avoir été désagrégé et rendu cristallin par un chauffage préalable assez prolongé. Après cela, il suffit de le porter au rouge et de le maintenir dans un mélange à teneur convenable de gaz et d'air, pour qu'il demeure incandescent. La température, à laquelle il se maintient ainsi spontanément, est bien inférieure à celle de sa fusion; cependant ce métal se sublime, passe dans la flamme qui se trouve au-dessus de lui et se dépose sur les objets qu'on y place. Fait singulier! le dépôt métallique ne se fait pas indifféremment sur la surface des objets, mais en des points particuliers correspondants à des saillies; alors même que celles-ci sont microscopiques, le dépôt métallique ensuite rigoureusement la trace.

Le mécanisme de ce dépôt est, à n'en pas douter, celui des dépôts métalliques effectués sur les pièces céramiques dites flammées.

A mon sens, ce phénomène doit avoir une portée plus générale: il expliquerait les dépôts et les effets de fusion et de scorification observés sur les revêtements réfractaires des fours à haute température, de même que la coloration de certains minéraux d'origine ignée.

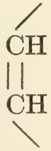
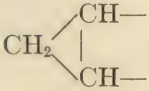
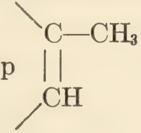
Il semble aussi qu'il expliquerait simplement certains faits constatés dans l'atmosphère du soleil, mais ce sujet demande de nouvelles études.

NEW RESEARCHES ON THE LUBRICATING OILS

By Prof. A. M. Nastukoff
Moscow, Russia, Arbat, Nikolsky, 10, 28

At the last Congress of Applied Chemistry in London[1] in 1909 I had the honour to communicate my essays on the condensation of the naphtha distillates with formalin in presence of less sulphuric acid than is necessary for obtaining ordinary solid and insoluble formolit. I have prepared in this manner from the fraction of petroleum, boiling at 200-250° C. and giving 12,8 as formolitic index, a fraction of condensed liquid hydrocarbons boiling at 186-188° C. at 50 mm. pressure and giving 105 as formolitic index. The later investigation[2] of this fraction has shown that there was the hydrocarbon $C_{15}H_{28}$; its specific gravity has been found $d^{21,5}/_{21,5} = 0,8498$ and index of refraction $n_{23} = 1,4703$; jodine value $= 0$.

The most characteristic feature of this quite new kind of cyclic hydrocarbons is the absence of the double bonds, th. i. they are saturated, but at the same time they give a very high formolitic index. The explanation I have done was following.

In the reaction take part the unsaturated groups  of the cyclic hydrocarbons and form with formalin trimethylene ring  ; the last get decomposed by strong sulphuric acid in presence of formalin in unsaturated group  capable of giving insoluble formolit.

I have prepared afterwards the same hydrocarbon from the fraction of petroleum boiling at 150-200° C.

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[1] A. M. Nastukoff. The Chemical Composition of the Lubricating Oils. Proceedings of the Seventh International Congress of Applied Chemistry, 1909.

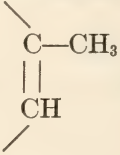
[2] A. M. Nastukoff and K. L. Maljaroff. "Trudy" of the Imperial Russian Technical Society in Baku, 1911. *Journal of the Russian Chemical Society*, 1910, p. 1596.

There is the short review of my published works in the matter since the last Congress in London. Now I have applied the same method of condensation to the fraction of petroleum, boiling at 250-300° C., in purpose to prepare new hydrocarbons of the series above mentioned. The preliminary essay has shown that the condensation goes as well as before on the other fractions. The formolitic index of the fraction itself was 18,65 and such of the residual left after distillation till 325° C.—46; the formolitic index of the like residual becomed after condensation was already 126.

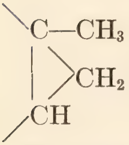
425 cc. of such a residuum gave me by distillation in vacuo at 220 mm. pressure till 339° C. the lower distillates with formolitic index 4, 10, and 22; and the asphaltum residue in the distilling flask giving 123 as formolitic index. The last was dissolved in benzol, the solution was filtered and distilled to get rid of the solvent. When the benzol ceased to come over, I have used the distillation of the residual by means of a current of superheated steam and obtained 18 gr. of a heavy oil of following properties: specific gravity $d^{1F}/_{1Z}=1,0113$; formolitic index 130; molecular weight 294; jodine value 20, 46; refactive index $n_{23}=1,5769$; viscosity — such of the cylinder oils.

The most characteristic feature of these condensed hydrocarbons is the high jodine value, by which they differ from those of the above-mentioned series.

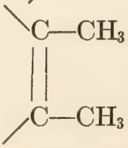
I suppose in this reaction the transition of the before mentioned reacting groups one in another taking place from beginning itself of the reaction. The group



get therefore in the group



and the last in



Here stops the condensation.

To sum up, the fraction of petroleum boiling at 250-300°C. was methylated by the formalin and gave in little amount the very true cylinder lubricating oil.

INFLUENCE DE LA GROSSEUR DU GRAIN DANS L'ANALYSE DES CHARBONS

Par le Capitaine P. Nicolardot
Directeur du Service des laboratories de l'artillerie

A Altofts on a constaté que les parties fines du charbon broyé n'ont pas la même composition que les parties les plus grossières. Les particules passées au tamis N° 100, et refusées au tamis N° 150, ont une humidité inférieure de 0,1%, une teneur en matières volatiles supérieure d'environ 3%, et une teneur en cendres inférieure d'environ 2%, aux valeurs correspondantes obtenues avec la partie traversant le tamis N° 240. Cette différence n'est pas un simple effet de la finesse, car les particules grossières pulvérisées à leur tour ne perdent, par cette opération, qu'une faible partie de leur teneur en matières volatiles et leur teneur en cendres augmente à peine.

Avec le charbon de Lievin, Monsieur Taffanel a constaté des différences de même genre dans les teneurs en matières volatiles et en cendres, mais en sens opposé. Ce fait tiendrait à ce que les impuretés stériles sont plus dures que le charbon.

Ces observations sembleraient devoir rendre illusoires toutes les précautions dont on s'entoure pour préparer les échantillons d'essai. Suivant que les opèrateurs opèrent sur des grains de grosseur différente, ils obtiennent, en se plaçant par ailleurs, dans des conditions indentiques, des résultats discordants. Quelques essais préliminaires exécutés sur des échantillons de charbons pris au hasard ne m'ont pas paru confirmer pleinement les conclusions des laboratoires d'Altofts et de Liévin. La teneur en matières volatiles a été déterminée par la méthode de Muck.

	Mailles	Par cmc	des tamis	
	225 .—	400 .—	870	1482
Anthracite	7,83	7,75	7,80	7,60
Houille N° 1	18,70	18,76	18,78	18,60
Houille N° 2	38,00	38,00	38,07	37,70

Les résultats obtenus avec les parties plus fines, quoique peu différents, sont cependant systématiquement inférieurs aux autres. J'ai cherché à opérer sur des échántillons de provenance certaine; et j'ai séparé au moyen des tamis renfermant par centimètre carré les nombres de mailles suivants:

16-25-56-100-225-650-992-1482-,

les divers échantillons en neuf portions différentes. La portion N° I était refusée par le tamis à 16 mailles, la portion N° 2 était constituée par les grains traversant le tamis à 16 mailles mais refusés par le tamis à 25 et ainsi de suite jusqu'à la dernière portion N° 9 constituée par toute la poussière et les grains traversant le tamis à 1482 mailles.

Pour déterminer la teneur en matières volatiles des diverses portions, j'ai opéré dans des conditions aussi identiques que possible en pesant des prises égales dans des creusets de porcelaine tarés et munis de leur couvercle. La méthode Muck et même la méthode de l'Ecole Supérieure des Mines de Paris, beaucoup plus précise encore, ne m'ont nas paru présenter assez de garanties de précision. Les essais ont été exécutés d'une part par la méthode de Phillips (chauffage pendant deux minutes sur un bec Bunsen, puis sur un chalumeau, toujours dans les mêmes conditions, pression du gaz, de l'air), et d'autre part en plaçant les creusets dans un moufle maintenu toujours à la même température. Les creusets étaient placés par trois dans le centre du moufle et laissés le même temps. Pour obtenir une plus grande régularité, les essais étaient alternés: tantôt ils étaient placés dans l'ordre naturel 1,2,3-4,5,6-7,8,9 ou autrement. Les résultats obtenus avec une anthracite du pays de Galles, un Cannel-Coal de New-Battle (Ecosse), des houilles de Courrières, Commentry, Decazeville et un lignite de Gardanne ont été les suivants:

	Portion N°	1	2	3	4	5	6	7	8	9
Anthracite Gallois	Eau	1,58	1,6	1,59	1,54	1,6	1,62	1,75	1,79	1,6
	Matières volatiles	7,00	7,1	6,95	6,97	7,1	7,0	6,98	6,95	6,96
	Cendres	3,8	3,87	3,83	3,9	3,87	3,95	4,0	3,9	3,85
Commentry	Eau	2,9	2,93	2,97	2,86	2,71	2,55	2,50	2,3	2,16
	Matières volatiles	34,95	35,00	34,9	34,85	35,00	34,87	34,80	34,6	34,5
	Cendres	4,0	3,98	3,8	3,78	3,82	3,95	3,85	3,95	4,0
Courrières	Eau	1,44	1,35	1,3	1,2	1,0	1,0	0,8	0,85	0,9
	Matières volatiles	29,00	29,5	29,1	29,0	28,9	29,1	29,1	28,9	28,8
	Cendres	2,7	2,8	2,9	2,7	2,2	1,2	2,3	3,4	3,1
Decazeville	Eau	6,14	6,82	6,81	6,8	6,77	6,76	6,77	6,7	6,42
	Matières volatiles	33,4	33,2	32,6	31,7	31,0	30,7	30,6	30,4	29,5
	Cendres	9,3	8,1	8,6	10,0	10,5	11,5	12,5	13,4	16,5
Cannel-coal de New-Battle	Eau	1,5	1,7	1,7	1,7	1,6	1,5	1,4	1,3	1,4
	Matières volatiles	51,0	50,0	48,0	47,0	46,0	45,0	43,7	43,5	40,0
	Cendres	3,85	3,6	3,5	3,7	3,6	3,3	3,2	4,2	4,8
Lignite de Gardanne	Eau	11,35	11,5	11,6	11,7	11,7	11,6	11,55	11,55	11,54
	Matières volatiles	40,5	41,0	41,3	41,4	41,1	40,0	40,1	40,0	39,9
	Cendres	3,3	3,4	3,2	3,55	3,45	3,4	3,4	3,3	3,4

Pour m'assurer que les variations observées n'étaient pas dûes au broyage, comme l'avait déja remarqué le Laboratoire d'Altofts, j'ai porphyrisé la partie N° 5 le l'échantillon de Cannel-Coal, qui présentait les plus grands écarts et dosait dans des conditions aussi identiques que possible l'eau, les cendres et les matières volatiles.

	Fraction 5 non porphyrisée —		Fraction 5 porphyrisée	
Eau	1,55	1,63	1,3	1,4
Matières Volatiles	46,05	45,85	45,85	45,65
Cendres	3,6	3,55	3,6	3,62

Des essais sur d'autres fractions du même Cannel Coal ou de la houille de Decazeville m'ont fourni des résultats analogues. L'action de broyage ne modifie pas sensiblement la composition des charbons et les variations dans leur teneur en cendres ou en matières volatiles, suivant que le grain est plus ou moins gros, tiennent à la nature même des différentes parties constitutives des charbons.

D'autres propriétés varient naturellement: le pouvoir calorifique s'abaisse dans les fractions les plus fines. La densité elle-même varie, quoique légèrement.

Fractions N°	1	2	3	4	5	6	7	8	9
Cannel Coal D	1,23	1,23	1,24	1,24	1,27	1,29	1,3	1,36	1,37

Je me propose de poursuivre ces recherches qui tendraient à montrer que la grosseur des grains doit être comprise entre certaines limites pour rendre comparables les résultats obtenus par divers opérateurs.

THE CONTINUOUS PURIFICATION OF COAL GAS WITH WEAK AMMONIA LIQUOR

By J. G. O'Neill
Geneva, N. Y.

In the manufacture of coal gas, the crude gas from the retorts is pulled by an exhauster through a condenser to free it from tar and considerable ammoniacial liquor, and is then forced by the exhauster into another condenser to rid it of excess heat, then through scrubbers which wash out the ammonia, and finally through the purifying boxes where it is purified from hydrogen sulphide and cyanogen, on into the holder. In washing the ammonia from the gas two scrubbers are used, the first scrubber, taking the gas, washes it with gas liquor, which absorbs a small amount of ammonia and hydrogen sulphide from the gas; the second scrubber washes the gas with fresh water which completely absorbs the ammonia from the gas.

The present practice of purifying coal gas from hydrogen sulphide and cyanogen consists of passing the gas through a mixture of iron oxide and shavings in air-tight boxes, whereby the hydrogen sulphide and cyanogen in the gas combine with iron oxide to form iron sulphide and ferrocyanides. When the iron oxide no longer takes up any hydrogen sulphide, it is taken from the boxes and exposed to the air, whence by oxidation and throwing off of free sulphur, it again becomes active or "revivified." This operation is repeated four or five times until the percentage of free sulphur in the purifying material increases to about 30%, when the iron oxide will no longer revivify. In earlier times the oxide would revivify and could be used until it contained 60% free sulphur, but with the modern practice of making gas at much higher temperatures the cyanogen in the gas has greatly increased, and a large percentage of the iron goes to form inactive ferrocyanides. By introducing about 2% of air into the gas before the purifiers, a certain amount of revivification takes place in the boxes, and reduces the number of times necessary to expose the oxide to the air to again make it active.

There are many disadvantages which we encounter in the iron oxide method of purification, some of which are as follows: the process is non-elastic, consequently, the normal increase of gas consumption from year to year; the very much greater consumption during certain periods of the year; the difficulty of obtaining coal low in sulphur, — all these conditions are forcing us to rapidly increase our purifying capacity at a very large expense, especially considering the facts that the purifying boxes are extremely bulky and that many coal gas plants are already cramped for space. The danger of a gas explosion every time a box is opened, and the close attention necessary to prevent firing of the oxide when exposed to the air for revivification. The cost of oxide and the labor cost in filling and emptying boxes, also the accumulation of spent oxide which gradually becomes a nuisance.

All these disadvantages of oxide purification have been recognized in the past, several investigators have devised other purification processes, none of which have met with general approval. Of these attempts, two are of interest to us, the first by F. C. Hills (British Patent No. 1369 of 1868 and 934 of 1874) whereby he heats ammonia liquor in a special apparatus to 180° F. and obtains a liquor having caustic properties. He washes the unpurified coal gas with this liquor and absorbs the hydrogen sulphide from it. His process failed because of the loss of ammonia in preparing the caustic liquor and because of his failure to determine the correct temperature of the liquor and gas during the washing. Had he determined this correct temperature, he would have found that double the quantity of ammonia from a ton of coal would absorb all the hydrogen sulphide in the gas from a ton of coal, instead of 6 to 7 times the ammonia as is believed to the present time.

The other process by C. F. Claus (U. S. Patent No. 337, 246 of 1886) also sought to use the caustic properties of ammonia. He prepared pure ammonia gas in an elaborate apparatus and introduced the pure ammonia gas directly into the unpurified coal gas, before the scrubbers. This process was fundamentally wrong, it complicated gas purification and increased its cost.

Any advance in the art of gas purification must have for its basis simplicity of operation. It must be more economical of labor, material and of space than the present oxide purification practice, and to be generally adopted it must be applicable to both the large and small coal gas plant. Such a process we have in operation in our plant for the past six months. Previous papers describing my process will be found in the *Progressive Age* of July, 1911 and August 15, 1911. In this article I wish to lay before you the manner of operation and the results obtained in the use of the process for the past six months.

In my method of operation I absorb the hydrogen sulphide, cyanogen and some carbon dioxide from the unpurified coal gas by means of a weak alkaline ammonia solution, prepared in an ordinary crude ammonia concentrator. I simply increase the quantity of weak ammonia liquor feeding into the ammonia still and draw off this increase of liquor near the outlet of the first ring, whose contents have been subjected to the action of steam containing a high percentage of ammonia vapor. We do not interfere with the actual amount of liquor being distilled or with the operation of the ammonia still. The liquor drawn from this ring is cooled to at least 30 degrees C. and is pumped into the first scrubber taking the place of the usual gas liquor wash. The only additional apparatus needed to carry out this process is a cooling apparatus which consists of a cooling coil with water spray and a small tubular cooler, which cools the liquor coming from the treatment ring of the ammonia still, thus cooling it for use in the first scrubber. The impurities given off by this liquor in the ammonia still saturate the concentrated ammonia in the condenser of the ammonia still, while the excess impurities pass off into the air after being washed of all traces of ammonia in the catch-pot.

The effects of washing the crude gas with this liquor are shown by the following data:

		Nov.11	Nov.22	Dec.5	Dec.7	Dec.14	Jan.2
Gas liquor to ammonia still	GPL NH_3—	7.00	5.27	4.25	3.87	6.15	7.00
	GPL H_2S —	3.20	2.49	1.98	2.31	3.57	3.20
Wash liquor from ammonia still	GPL NH_3 —	7.48	6.93	5.50	4.79	5.98	8.48
	GPL H_2S —	1.46	1.12	.68	.74	.80	1.46
Wash liquor from first scrubber	GPL NH_3 —	7.48	6.83	5.84	5.03	7.82	9.18
	GPL H_2S —	6.12	6.00	3.80	4.43	6.13	8.17
Grains H_2S per 100 cu. ft. in crude gas before scrubbers		807.8	703.8	592.1	703.8	557.0	966.9

	Nov.11	Nov.22	Dec.6	Dec.7	Dec.14	Jan.2
Grains H_2S per 100 cu. ft. in crude gas after scrubbers	276.4	96.3	33.6	44.0	25.0	30.6
No. of thousand cu. ft. gas washed per hour	45	40	35	37	45	44
Gallons liquor wash used per hour	720	815	700	900	600	900
Grains H_2S eliminated per gallon liquor wash used	314	298	279	270	399	574

All of the cyanogen in the crude gas is eliminated by the liquor wash.

On an average, I found the liquor wash absorbed from 0.3 to 0.5% of carbon dioxide from the gas. My opinion is this absorption would be greatly increased with a liquor wash containing a higher percentage of ammonia and with a longer time of contact.

The temperature of the liquor wash to the first scrubber must not be higher than 30 degrees C. neither should the temperature of the gas to the scrubber be higher than 30 degrees C. The best results are obtained when the two temperatures are held at about 25° C. This is the temperature of the gas to the scrubbers that we meet with in ordinary coal gas practice. If gas liquor is substituted for the liquor wash there is practically no increase in hydrogen sulphide in the gas liquor or decrease of hydrogen sulphide in the gas.

Since our scrubbing apparatus was limited to one fresh-water scrubber consisting of eight bell-shaped compartments, I divided the scrubber using the four lower compartments for the liquor wash and the four top ones for fresh water. The results would have been much more satisfactory if I had one scrubber for the liquor wash and one for the fresh water, then I could have used much stronger liquor and would have obtained much higher elimination of impurities per gallon of liquor wash used, without fear of losing any ammonia from insufficient washing.

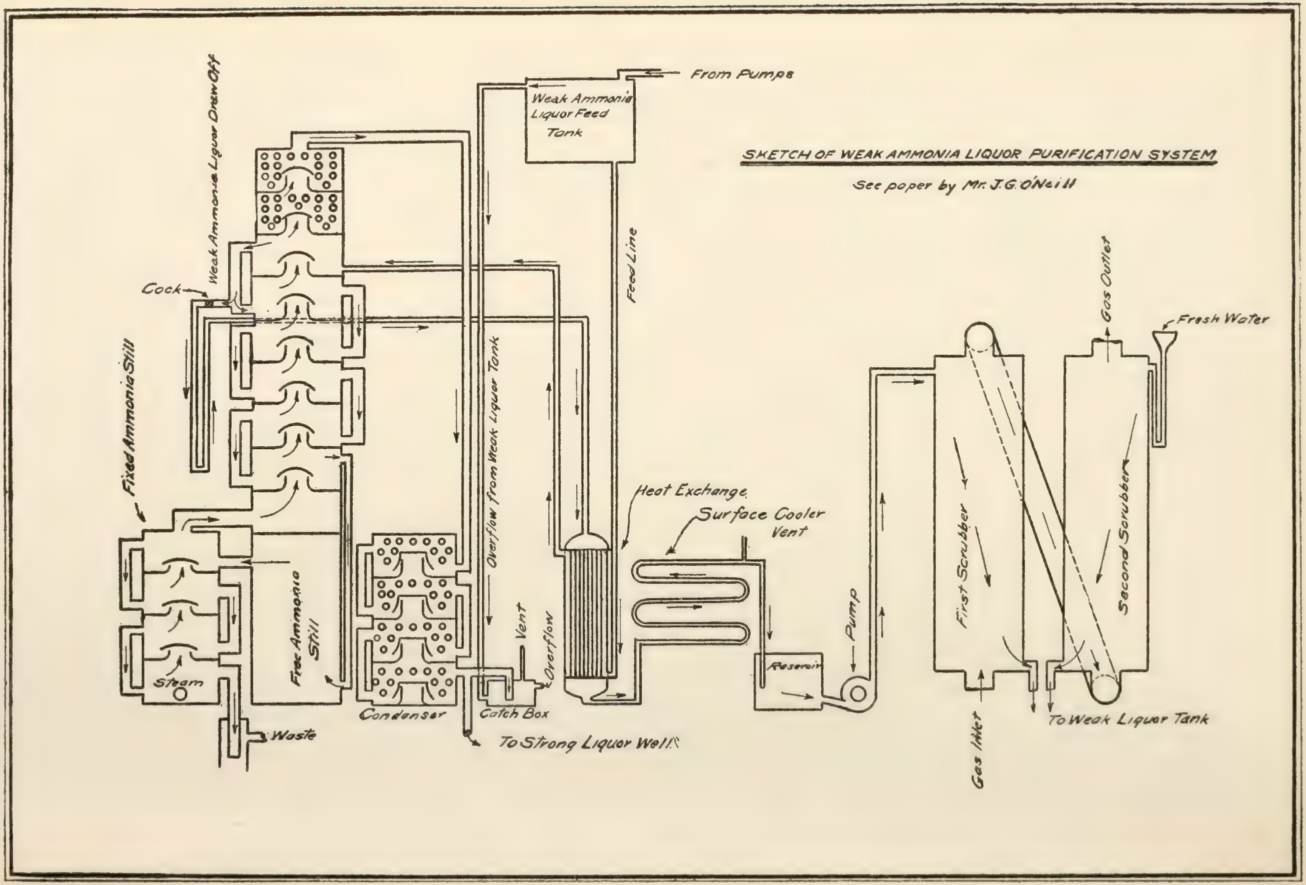
Another very disagreeable feature of our operation was the uneven make of gas, the amount of gas washed for one hour differing as much as 50% from another hour of the same day, with little regulation of the liquor wash to meet the increase or decrease in the gas.

Considering all the disadvantages mentioned above still nowhere in the process was there the least possibility of ammonia loss, also the average yield of ammonia for the six months exceeded the average yield for the six months previous to the installation of the process. No difficulty was experienced in the operation of the ammonia still. The total amount of steam required to operate the ammonia still was not appreciably increased because the cooling water for the ammonia vapors was practically eliminated.

During the month previous to installing the process, 27% of the coal used for gas making was a high sulphur coal. The purifying capacity consisted of three boxes, holding a total of 6000 bushels of shavings and iron oxide, during this month we found it impossible to change the oxide in the boxes rapidly enough to purify the gas from hydrogen sulphide, even using new oxide and introducing some air into the gas before the purifiers.

For the six months after installing the liquor purification process, the oxide in the boxes was not changed, no labor was expended on purification, the average monthly output of gas for the first three months increased 12% and for the last three months decreased 20% (partly due to coal shortage), the percentage of high sulphur coal used was increased to 50% of the total, and at the end of this period we found all boxes clean. A very small quantity of air was drawn into the gas before the exhausters, approximately 0.5%; the quantity of nitrogen in this air introduced into the gas, was more than offset by the quantity of carbon dioxide absorbed from the gas by the liquor wash. The gas issuing from the first purifying box in the series would at times show a trace of hydrogen sulphide, but on changing the flow of the gas, so as to remove this box from being first to receive the gas, it would immediately clean up. The gas has gone through the boxes for a month at a time without the gas from the first box showing a trace of hydrogen sulphide with lead acetate paper.

During the severe winter weather, the wash liquor in the pipe line to the scrubber became frozen, consequently for several hours the liquor wash was discontinued. The three boxes went foul even though a short time before they were all perfectly clean. When the wash liquor was again turned on the three boxes grad-



SKETCH OF WEAK AMMONIA LIQUOR PURIFICATION SYSTEM

See paper by Mr. J. G. O'Neill

ually cleaned up, proving that by using this liquor wash with a very small quantity of air, the foul boxes could again be made clean, without the necessity of first passing the gas through a clean box.

The actual saving obtained by using this process is very large, even for a small plant, both in labor cost and using high sulphur coal; the process is simpler than oxide purification, elastic, independent of one grade of coal, no danger of fire or explosions, does away with any installation of new purifying boxes and saves valuable space.

The gas business being a world-wide industry, consequently any improvement in the manufacture of gas is of a world-wide interest. With the modern development of uses for gas, any improvement in its manufacture is a direct aid to the progress of the world.

CALCIUM CARBONATE AS A CONSTITUENT OF COAL ASH[1]

By S. W. Parr
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Calcium carbonate as a constituent of coal is recognized without question as a disturbing element in the determination of some of the most important data pertaining to this material. In the ultimate method of analysis it augments the factor for total carbon by reason of the CO_2 disengaged in that determination. Similarly the ash value as weighed is too low and the oxygen factor, determined as it is by difference, is too high. In the calculation of heat values, therefore, according to the DuLong formula the values are in error both on account of the total carbon factor and also because of the value indicated for available hydrogen. These errors partly neutralize each other, it is true, but opportunity would still remain for introducing considerable of a discrepancy into the calculation. In the proximate method of analysis also, too high a factor would be indicated for the volatile matter resulting from the decomposition of the calcite. It would thus appear that the CO_2 present would be reckoned by the engineer as "fuel" when calculating his efficiencies to the "ash and water free basis." For these reasons, therefore, it is strange that this ingredient has not received earlier attention.

In the report of the committee of the American Chemical Society on Coal Analysis[2] the question was asked: "Are carbonates likely to be present in the ash in such amount that heating over a blast lamp will lessen the weight appreciably?" Probably the first serious attempt to answer this question has been taken up recently in a study of the composition of the ash of Illinois coals. The first results of this study were published as part of Bulletin No. 37 of the University of Illinois Engineering Experiment Station.[3] The data therein contained covered a range of 50 coals from 15 counties. The results showed calcium carbonate to be uniformly present, but in widely varying amounts. The studies were continued in order to determine if the distribution of this ingredient, or the increase or decrease of the same in quantity, conformed to any well-defined areas. Fifty additional samples were therefore examined and the results published in Bulletin No. 16 of the Illinois State Geological Survey. These studies have been continued in connection with the work of the survey until at the present time the total samples examined number 235, coming from nearly every coal-producing county in the State.

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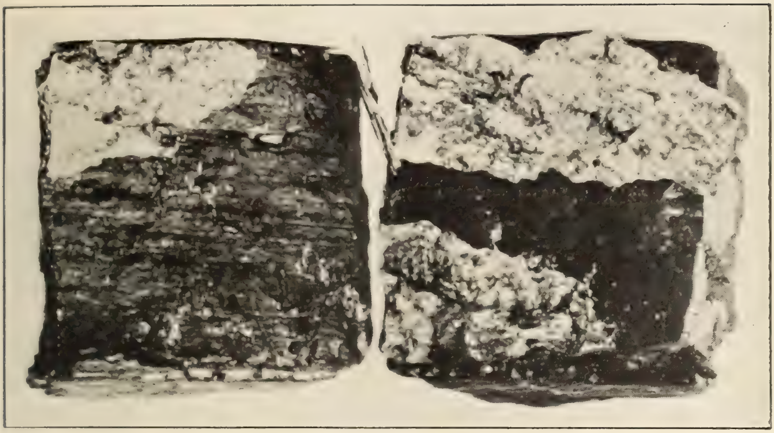
[1] *Journal American Chemical Society*, Vol. 20, p. 284, 1898.
[2] Coal Studies in connection with the Illinois State Geological Survey.
[3] Unit Coal and the Composition of Coal Ash. By S W. Parr and W. F. Wheeler, 1909.

In discussing this constituent it may be well at the outset to refer to the form in which it occurs. It has been a common belief that the white flakes frequently observed in the cleavage planes of coal were calcium sulphate. An examination of such material somewhat frequently made by the writer found it in fact to be gypsum. However, the analytical data above referred to, showing an almost universal distribution of calcium carbonate in coal, have led to a more critical examination with the result that not infrequently the white scales are found to be calcite. These two substances, moreover, differ in appearance to such an extent that they can usually be readily distinguished without resort to chemical or microscopic examination.

Figure 1, for example, illustrates a quite liberal flaking in the cleavage planes of calcium sulphate. In appearance it is a "dead" or "flat" white without lustre.

Figure 2 shows a layer of calcite. It is much more vitreous in appearance with the suggestion that it might be semi-transparent if held up to the light. And upon trial it is found to have this property.

In the accompanying table the CO_2 is given in percentages of the dry coal. It is also calculated to calcium carbonate and given in percentages both of the dry coal and of the standard ash. It should be noted, moreover, that these samples were taken at the working face of the vein, crushed and quartered according to standard methods, sealed in tins and sent direct to the laboratory. Certain considerations seem to be suggested by the results as shown. The occasional excessively high percentage of CO_2 is striking and indicates the need for taking account of this con-



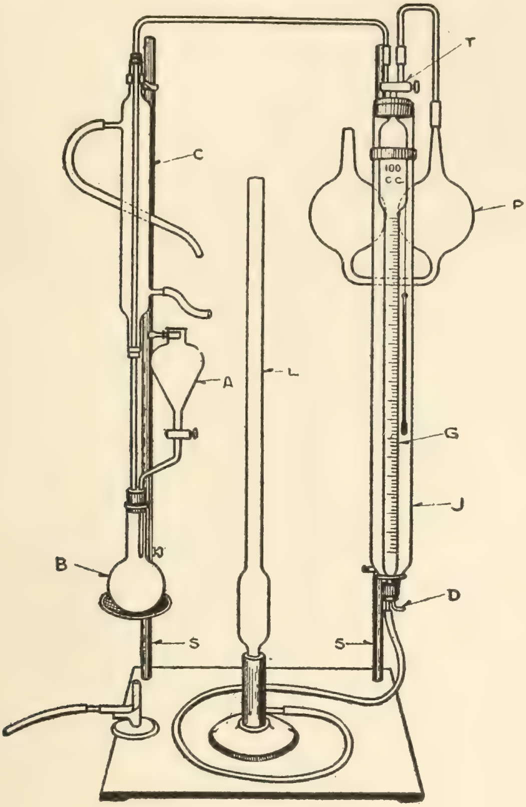


Figure 3

stituent in all coals where the ash content is high enough to admit of so large a disturbing factor. For example, in two cases, one in Bureau County and one in Franklin County, the content of CO_2 exceeds 4 per cent, representing a calcium carbonate factor of over 10 per cent of the weight of the coal. Again five samples exceed 2 per cent of CO_2, representing in every case an amount of calcium carbonate in excess of 5 per cent of the total weight. There are also 36 samples with more than 1 and less than 2 per cent of CO_2. That is to say, over 15 per cent of the 235 samples have an amount of carbon dioxide in excess of one per cent. And if we count in all samples having a sufficiently high content of this material to represent a content of calcite in excess of 2 per cent the number reaches 55 in 235 samples, or over 23 per cent.

In view of the ease with which this constituent can be measured it should not be considered so much of a burden as to make its determination objectionable. The apparatus involves no essentially new features but is a simple and convenient assembling of well known units.

Figure 3 illustrates the apparatus used throughout these experiments. Five grams of powdered coal are placed in the flask B and acid is added from the dropping funnel A. At the close of the operation, after boiling out the CO_2 from the solution, the flask and capillary are filled with water to the three-way cock T. The carbon dioxide is measured by difference after absorption in the potash solution in P.

So far as the data in hand are concerned, the most important generalization regarding the distribution of calcite in the coal measures would seem to be that its occurrence is entirely adventitious and bears little if any relation to the overlying material. The same region or county and even the same mine will frequently show variations almost as widely apart as the extremes throughout the State. One possible exception may be noted. In Perry County the range of variation is relatively narrow and the values rarely exceed 0.5 per cent of CO_2.

			In Percentage of Dry Coal			
No.	County	Seam	Ash as determined by the standard method	CO_2	CO_2 calculated to $CaCO_3$	Per ct. of $CaCO_3$ in ctd. ash
617	Bureau	2	11.62	.94	2.13	13.8
2621	Bureau	6	20.38	4.46	10.12	36.8
2622	Bureau	6	10.61	.52	1.18	9.7
2634	Bureau	6	12.77	.98	2.22	13.1
4389	Bureau	2	9.07	.68	1.55	12.04
4390	Bureau	2	8.46	.81	1.84	14.83
2641	Calhoun	2	12.97	.62	1.41	7.9
2811	Christian	6	12.01	.56	1.27	7.7
2784	Christian	6	11.16	1.20	2.72	17.3
2785	Christian	6	10.61	.64	1.45	9.9
2798	Christian	6	9.18	.07	.16	1.3
996	Clinton	6	10.47	.44	1.00	6.9
1176	Clinton	6	13.59	.40	.91	5.1
1178	Clinton	6	15.56	2.48	5.63	26.9

No.	County	Seam	In Percentage of Dry Coal			
			Ash as determined by the standard method	CO_2	CO_2 calculated to $CaCO_3$	Per ct. of $CaCO_3$ in ctd. ash
2635	Clinton	6	10.76	.28	.63	5.0
2636	Clinton	6	12.99	.80	1.82	10.7
419	Franklin	6	10.11	.37	.84	7.2
420	Franklin	6	7.53	.21	.48	5.4
461	Franklin	6	——	.28	.63	——
4381	Franklin	6	7.75	.35	.79	8.10
4382	Franklin	6	10.27	.49	1.11	8.55
4782	Franklin	6	5.69	.09	.21	3.22
4783	Franklin	6	3.31	.05	0.11	2.70
4784	Franklin	6	11.98	.54	1.23	7.97
4785	Franklin	6	14.51	4.89	11.10	52.68
4786	Franklin	6	6.96	.36	.82	9.90
4787	Franklin	6	7.14	.25	.57	6.85
4788	Franklin	6	4.19	.01	.02	.42
4789	Franklin	6	7.75	.20	.45	5.08
4790	Franklin	6	9.71	.25	.57	5.20
4791	Franklin	6	8.49	.34	.77	7.79
4792	Franklin	6	7.30	.61	1.38	15.63
4793	Franklin	6	8.53	.25	.57	5.77
4794	Franklin	6	8.53	.41	.93	9.14
4809	Franklin	6	10.40	.37	.84	5.99
4810	Franklin	6	11.24	.99	2.25	15.11
4811	Franklin	6	11.14	1.53	3.47	22.40
4812	Franklin	6	10.63	1.28	2.91	19.92
4813	Franklin	6	11.17	.76	1.72	10.15
1404	Fulton	5N	12.52	1.30	2.95	17.4
2648	Fulton	5N	11.35	1.02	2.31	15.3
2751	Fulton	5N	15.59	1.43	3.25	15.3
2752	Fulton	5N	13.05	1.01	2.29	13.0
2753	Fulton	2	6.40	.09	.20	2.2
2757	Fulton	5N	12.40	1.32	3.00	1.76
2758	Fulton	5N	11.55	1.01	2.29	14.3
4387	Fulton	5N	13.65	.82	1.86	10.5
4388	Fulton	5N	12.27	.65	1.47	5.12
1092	Gallatin	5S	10.85	.72	1.63	11.2
2750	Greene	6	11.37	.64	1.45	9.6
743	Grundy	2	8.17	.15	.34	3.1
734	Grundy	2	5.82	.88	1.98	24.2
2609	Grundy	2	13.01	.30	.68	3.8
2624	Grundy	2	12.88	3.07	6.97	38.5
4391	Grundy	6	6.79	.39	.89	9.59
4392	Grundy	6	6.73	.47	1.07	11.63
2630	Henry	6	12.38	.99	2.25	13.6
2776	Henry	3	12.53	1.70	3.86	24.8
2777	Henry	3	12.13	1.73	3.92	22.0
2737	Jackson	2	5.78	.18	.41	5.9
2738	Jackson	2	6.41	.15	.34	4.2
2754	Jackson	6	7.66	.11	.25	2.7
2755	Jackson	6	10.55	.05	.11	.8
4771	Jackson	6	5.00	.93	2.11	31.59
4772	Jackson	6	3.60	.06	.14	3.19

			In Percentage of Dry Coal			
No.	County	Seam	Ash as determined by the standard method	CO_2	CO_2 calculated to $CaCO_3$	Per ct. of $CaCO_3$ in ctd. ash
4773	Jackson	6	9.55	.34	0.77	6.97
4780	Jackson	6	12.08	.47	1.07	6.76
4781	Jackson	6	8.83	.39	0.89	7.25
2756	Knox	3	7.71	.06	.14	1.3
2769	Knox	6	10.48	.84	1.91	13.8
2652	LaSalle	2	5.91	.19	.43	5.2
2612	Livingston	5N	14.63	.45	1.02	5.5
2804	Macon	6	11.47	.49	1.11	7.2
735	Macoupin	6	10.86	.36	.82	5.4
736	Macoupin	6	11.4	.13	.29	1.9
2786	Macoupin	6	10.21	.31	.70	5.0
2788	Macoupin	6	10.11	.36	.82	5.9
2818	Macoupin	6	12.00	.11	1.25	1.7
2819	Macoupin	6	11.56	.53	1.20	7.4
1117	Madison	6	10.59	.64	1.45	10.1
1118	Madison	6	13.65	.55	1.25	7.4
1119	Madison	6	11.72	.71	1.61	9.9
2608	Madison	6	11.54	.60	.45	2.7
2653	Madison	2	12.29	.42	.95	5.6
2816	Madison	6	12.27	.18	.41	2.5
2817	Madison	6	11.88	.20	.45	2.9
2614	Marion	6	10.89	.31	.70	4.7
2627	Marshall	2	8.26	.41	.93	8.4
2779	Menard	5N	12.57	.87	1.97	11.4
2782	Menard	5N	10.86	.84	1.86	12.1
2783	Menard	5N	11.18	.88	2.00	12.7
4383	Menard	5N	9.73	.83	1.88	14.13
4384	Menard	5N	12.90	1.13	2.57	14.93
2650	Mercer	1	8.10	.81	1.83	15.1
2820	Montgomery	6	13.78	1.08	2.45	13.6
2739	Morgan	2	17.66	1.07	2.43	10.6
2812	Moultrie	6	12.04	.17	.39	2.4
1403	Peoria	5N	15.46	2.48	5.63	26.9
1407	Peroia	5N	16.25	2.15	5.33	24.4
1410	Peoria	5N	14.78	1.57	3.89	19.5
1411	Peoria	2	10.90	.98	1.79	11.7
2639	Peoria	5N	10.95	.79	1.79	12.3
2640	Peoria	5N	16.83	.59	1.40	6.7
2642	Peoria	5N	10.43	.67	1.52	11.2
2643	Peoria	5N	13.57	1.05	2.38	13.5
2644	Peoria	5N	12.64	1.02	2.31	13.5
421	Perry	6	14.71	.65	1.47	8.6
422	Perry	6	——	.28	.63	——
2615	Perry	6	8.17	.20	.45	4.6
2623	Perry	6	14.03	.46	1.03	5.7
2626	Perry	6	12.03	.54	1.22	7.9
4756	Perry	6	10.55	.26	.59	4.80
4747	Perry	6	10.09	.15	.34	2.94
4758	Perry	6	9.49	.25	.57	5.15
4759	Perry	6	11.54	.25	.57	4.30
4760	Perry	6	9.86	.25	.57	4.94

No.	County	Seam	In Percentage of Dry Coal			
			Ash as determined by the standard method	CO_2	CO_2 calculated to $CaCO_3$	Per ct. of $CaCO_3$ in ctd. ash
4761	Perry	6	11.53	.32	.73	5.46
4762	Perry	6	3.51	.14	.32	7.14
4763	Perry	6	3.40	.06	.14	3.24
4764	Perry	6	12.67	.37	.84	5.77
4765	Perry	6	13.30	.06	.14	0.94
4766	Perry	6	12.47	.39	.89	6.20
4767	Perry	6	12.83	.38	.86	5.83
4768	Perry	6	12.08	.70	1.59	11.10
4769	Perry	6	11.98	.31	.70	5.04
4770	Perry	6	7.60	.34	.76	7.77
2638	Putnam	2	8.81	.31	.70	5.9
2625	Randolph	6	11.52	.44	1.00	6.7
2775	Rock Island	1	9.87	.66	1.50	10.1
725A	St. Clair	6 Bottom	14.86	.70	1.59	7.2
725B	St. Clair	6 Middle	——	.72	1.63	10.3
725C	St. Clair	6 Top	——	.37	.84	8.9
991	St. Clair	6	15.80	——	——	——
1001	St. Clair	6	12.53	.55	1.25	8.1
1002	St. Clair	6	13.43	.50	1.13	6.7
1003	St. Clair	6	12.94	.76	1.72	9.8
2628	St. Clair	6	12.57	.36	.82	4.9
2631	St. Clair	6	12.46	.24	.53	3.2
2632	St. Clair	6	12.95	.34	.77	4.5
2633	St. Clair	6	12.79	.47	1.07	6.3
2637	St. Clair	6	12.91	2.58	5.86	30.9
4385	St. Clair	6	12.95	.54	1.22	7.12
4386	St. Clair	6	12.51	.38	.86	5.09
1094	Saline	5S	10.54	.26	.59	4.5
1095	Saline	5S	11.49	1.22	2.77	17.4
1110	Saline	5S	12.68	.51	1.16	6.6
1112	Saline	5S	8.99	.11	.25	1.9
1115	Saline	5S	11.58	.34	.77	5.2
1116	Saline	5S	9.89	1.51	3.43	25.4
1120	Saline	6	——	1.20	2.72	——
4379	Saline	5S	8.85	.13	.30	2.58
4380	Saline	5S	7.56	.14	.32	3.17
540	Sagnamon	5N	10.76	.32	.73	5.0
720	Sagnamon	5N	13.81	.54	1.22	7.0
722	Sagnamon	6	11.04	.19	.43	2.9
740	Sagnamon	5N	12.75	.34	.77	4.7
741	Sagnamon	5N	12.47	.85	1.93	11.5
2740	Scott	2	10.19	.64	1.45	10.8
2815	Shelby	6	14.49	.40	.91	4.9
2629	Stark	6	10.60	2.40	5.45	33.6
1412	Tazewell	5N	——	.61	1.38	——
1413	Tazewell	5N	——	1.36	3.09	——
557	Vermilion	6	9.15	1.08	2.45	20.9
558	Vermilion	6	8.03	.47	1.07	10.5
4378	Vermilion	6	12.32	.69	1.57	9.73
4668	Vermilion	6	10.23	.63	1.43	10.78
4669	Vermilion	6	9.40	.49	1.11	9.42

			In Percentage of Dry Coal			
No.	County	Seam	Ash as determined by the standard method	CO_2	CO^2 calculated to $CaCO_3$	Per ct. of $CaCO_3$ in ctd. ash
4670	Vermilion	6	10.05	.52	1.18	8.92
4671	Vermilion	6	9.54	.62	1.41	11.63
4672	Vermilion	6	10.66	.77	1.75	12.08
4673	Vermilion	6	10.40	.91	2.07	15.49
4674	Vermilion	6	8.98	.63	1.43	12.66
4675	Vermilion	6	8.96	1.11	2.50	21.42
4676	Vermilion	6	10.04	.86	1.95	16.41
4677	Vermilion	6	10.50	1.11	2.50	18.17
4678	Vermilion	6	10.15	1.08	2.45	18.53
4679	Vermilion	6	8.72	.46	1.04	9.63
4697D	Vermilion	6	18.73	1.69	3.84	15.58
4698D	Vermilion	6	16.79	.61	1.38	6.47
4699	Vermilion	6	11.36	.63	1.43	10.04
4700	Vermilion	6	13.82	1.23	1.79	10.06
4701	Vermilion	6	12.79	1.29	2.93	17.68
4702	Vermilion	6	13.52	1.15	2.61	15.12
4703	Vermilion	6	13.47	1.26	2.86	16.36
4704	Vermilion	6	11.66	.81	1.84	12.38
4705	Vermilion	6	8.57	.42	.95	8.18
4706	Vermilion	6	9.34	.84	1.91	14.88
4707	Vermilion	6	11.44	1.02	2.32	14.48
4708	Vermilion	6	10.56	.95	2.16	14.76
4710	Vermilion	7	11.29	.52	1.29	8.77
4711	Vermilion	7	11.27	.52	1.29	8.57
4712	Vermilion	7	10.46	.47	1.07	7.84
4713	Vermilion	7	10.15	.59	1.34	10.07
4714	Vermilion	7	11.56	.70	1.59	10.16
4715	Vermilion	7	11.76	.88	2.00	12.85
4716	Vermilion	7	12.47	.96	2.18	13.08
4717	Vermilion	7	11.03	.65	1.47	9.87
4718D	Vermilion	7	25.71	1.15	2.61	8.30
4719D	Vermilion	7	40.90	.46	1.04	2.18
4720D	Vermilion	7	48.56	.29	.51	.89
4721	Vermilion	7	10.33	.46	1.04	7.62
4722	Vermilion	7	11.00	.54	1.23	8.51
4723	Vermilion	7	10.95	.38	.86	6.00
4724	Vermilion	7	10.18	.44	1.00	7.41
4727	Vermilion	7	12.81	.80	1.82	11.06
4728	Vermilion	7	11.34	.74	1.68	11.62
4729D	Vermilion	6	41.51	.81	1.84	3.88
4730D	Vermilion	6	18.20	.81	1.84	7.63
4731D	Vermilion	6	47.59	1.66	3.77	6.66
4734	Vermilion	7	10.75	.64	1.45	10.60
4735	Vermilion	7	12.64	.83	1.88	11.59
4736	Vermilion	7	12.00	.53	1.20	7.94
4737	Vermilion	7	11.79	.62	1.41	9.48
4738	Vermilion	6	10.44	.55	1.25	9.33
4739	Vermilion	6	12.74	.56	1.27	7.91
4740	Vermilion	6	13.18	.96	2.18	12.66
4741	Vermilion	6	12.30	.90	2.04	12.72
4742	Vermilion	6	10.56	.52	1.18	8.75

			In Percentage of Dry Coal			
No.	County	Seam	Ash as determined by the standard method	CO_2	CO_2 calculated to $CaCO_3$	Per ct. of $CaCO_3$ in ctd. ash
4743	Vermilion	6	9.28	.39	.89	7.68
4744	Vermilion	6	9.38	1.14	2.59	20.98
4745	Vermilion	6	9.48	1.15	2.61	20.88
4746	Vermilion	6	12.94	.72	1.63	10.00
4753D	Vermilion	7	18.51	1.09	2.47	10.48
4754D	Vermilion	7	19.09	.89	2.02	8.44
2613	Washington	6	13.61	.42	.95	5.9
2612	Washington	6	10.33	.64	1.45	10.3
1121	White	6	11.50	.41	.93	6.1
2611	Will	2	6.44	.61	1.38	16.0
896	Williamson	5	10.68	.70	1.59	11.1
459	Williamson	6	8.48	.12	.37	3.7
460	Williamson	6	10.13	.47	1.07	9.7
462	Williamson	6	7.66	.14	.32	3.3
1088	Williamson	6	10.65	.35	.79	5.9
2649	Woodford	2	6.17	.61	1.38	17.6

THE RESINIC BODIES IN BITUMINOUS COAL[1]

By S. W. Parr

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By resinic bodies is meant those degradation products resulting from the decomposition and transformation by geologic processes of the original vegetable matter of the resinous type. The presence and character of this material is based on the fact that oxygenated compounds exist in coal which may be removed by certain solvents, as phenol, etc.; that they are unsaturated and may readily take up and unite chemically with oxygen; that they may be distilled at low temperatures yielding distillates that have the same characteristics as to avidity for oxygen and chemical union with the same; and upon extraction they are found to have a rather definite melting point and this, moreover, is below the temperature at which decomposition occurs. These characteristics are all of a pronounced type and are of such a nature as to impart to the coal itself the most fundamental of its characteristics. It is, therefore, upon a better understanding of the character and behavior of these bodies that our real knowledge of coal must be based. It is the purpose of this paper to call attention to some of the more important of the properties and effects produced by the presence of these resinic residues.

1. *The Deterioration of Coal Samples in Laboratory Containers.* This behavior is directly due to the avidity of the freshly mined coal for oxygen. The studies of Parr and Barker on the "Occluded Gases" in coal[2] show this characteristic in a marked degree and also that the oxygen is combined chemically and retained as part of the structure of the coal.

There is, moreover, direct evidence of deterioration of coal samples after preparation by air drying, grinding and storage in glass jars with rubber seal. The amount of deterioration varies with different varieties of coal and with the length of time in storage, but is not sufficiently uniform to be definitely expressed

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[1] Coal Studies in connection with the Illinois State [illegible]

[2] Bulletin No. 32, University of Illinois Engineering Experiment Station, 1909.

DETERIORATION OF COAL

Comparison of values obtained in 1908-1909, with values January, 1912

Lab. No.	Date of 1st determination	Description of Sample	Moisture in Sample		DRY COAL								Difference in BTU on unit Coal
					Ash		Sulphur		B. T. U.		Unit Coal Ash moisture and Sulphur free		
					I	II	I	II	I	II	I	II	
1758	8–08	Ill-No. 1	1.83	2.58	14.25	13.86	7.61	7.25	12010	11422	14573	13646	1027
1858	9–08	Ill No. 1	1.62	2.02	7.99	8.05	4.71	4.67	13464	12954	14904	14335	569
2650	9–08	Ill-No. 1	10.35	2.20	8.10	7.35	4.59	4.21	13311	12402	14474	13818	656
1765	8–08	Ill-No. 2	2.73	2.54	9.77	10.01	3.36	3.44	13033	12496	14686	14116	570
1775	8–08	Ill-No. 2	4.33	2.90	7.29	7.43	1.90	1.96	13539	13059	14761	14258	503
1761	8–08	Ill-No. 5N	3.87	3.08	12.48	12.67	4.55	4.35	12364	11841	14444	13849	595
1856	9–08	Ill-No. 5N	1.84	2.22	12.22	12.11	4.17	4.10	12666	12194	14673	14158	515
1092	2–08	Ill-No. 5S	1.42	1.69	10.85	10.66	3.72	3.27	13235	12615	15133	14362	771
1095	2–08	Ill-No. 5S	1.68	2.12	11.49	11.66	4.16	4.13	12931	12287	14856	14311	545
1776	8–08	Ill-No. 6	4.40	3.45	6.42	6.34	1.33	1.36	13620	13151	14626	14157	469
1841	9–08	Ill-No. 6	4.56	2.98	12.68	12.39	6.49	6.53	12214	10733	14356	12533	1823
1870	10–08	Ill No. 6	4.57	3.54	9.97	9.92	3.70	3.74	12745	12133	14405	13694	801
1784	8–08	Ill-No. 7	5.29	3.36	12.86	12.69	4.04	4.13	12637	11921	14742	13941	801
1843	9–08	Ill-No. 7	2.81	2.32	11.43	11.39	3.17	3.19	12842	12477	14680	14298	382
2180	2–09	Ill-Vermilion	2.28	2.64	22.92	22.79	5.19	5.02	10924	10430	14731	14014	717
2075	1–09	Oklahoma	3.05	.92	8.77	11.00	4.30	4.13	14110	13400	15761	15473	288
2140	2–09	Anthracite	1.04	1.54	13.27	13.16	1.21	1.25	13010	12813	15233	14982	251
2114	2–09	Ky-Fox-Ridge	1.31	1.52	8.73	8.73	4.47	4.34	13783	13256	15391	14802	589
2125	2–09	Ala. Blk. Crk.	1.92	.77	12.87	13.68	.94	1.14	13639	13408	15883	15781	102
2128	2–09	Md. Geos. Crk.	.78	.46	8.55	8.62	1.13	1.13	14343	14225	15852	15731	121
2113	2–09	Kawana Splint	1.12	1.24	15.11	15.08	.93	.83	12710	12672	15224	15168	.56
2127	2–09	Pa. Wash. Gas	1.53	1.45	8.45	8.40	.98	1.02	13938	13748	15374	15156	218
2132	2–09	Pa. Cambria	.64	.38	4.14	4.22	.70	.60	15241	15022	15971	15794	177
2137	2–09	Pa. Sugar Loaf	.66	.68	5.90	6.00	1.32	1.37	14795	14654	15857	15796	.61
2144	2–09	Pa. Floyd City	1.71	1.88	4.97	4.88	1.00	.94	14298	13944	15153	14751	402
3045	2–10	Wyoming	17.98	12.08	10.02	9.57	2.54	2.67	10973	10277	12352	11500	852

in percentage form. A loss from 3 to 5 per cent over a period of three years is a common factor for coals of the Illinois type. With Eastern coals the deterioration is more nearly within a range of from ½ to 1½ per cent.

With certain coals, especially in Illinois, there is an active deterioration during the period immediately after breaking out from the vein and which covers the time in transit and in process of preparation and grinding for calorimetric determinations. This loss ranges from ½ to 1 per cent in the first ten days.

While it is not possible to indicate any general rule covering the case, it is evident that comparative values in the matter of the heat determinations should refer to samples tested at approximately the same date. As illustrating this fact of deterioration there is given in the accompanying table the log of values on 25 coals which have been in storage for varying lengths of time.

2. *The Weathering of Coal.* This effect is again directly traceable to the avidity of the resinic bodies for oxygen. From studies on weathering in this laboratory[1] there is evidence that in the preliminary stages of weathering, that for example which occurs during the first year, the weathering effect is seemingly only that amount and kind of deterioration represented by the attraction of the resinic bodies for oxygen. In this connection it is interesting to note the substantial agreement in the loss of heat values sustained by coal masses in open air storage and the pulverized samples under conditions of laboratory storage.

3. *The Free Burning Property of Fresh Coal.* This property is doubtless largely due to the chemical activity of the resinic bodies. Coal that has been exposed to the air for some time, especially in the finer sizes, is said to be "dead" as to its behavior on the grates. Pillar coal from the mines behaves similarly and must be sold at a reduced price. The firemen who use such coal are of the opinion that it has lost a large per cent of its heat values. The active constituents however have become saturated with oxygen and while this slow reaction has only at the most reduced the heat value by 3 or 4 per cent it has seriously impaired the burning property of the fuel. The speed of combustion is so greatly reduced

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[1] Bulletin No. 38 University of Illinois Engineering Experiment Station, 1909. Parr and Wheeler.

it is said to be "dead" instead of "free burning" and in fact because of the greater draft required its real efficiency has been impaired to a degree much greater than would be indicated by the simple matter of loss in heat content. Lignites which are especially deficient in these more active resinic bodies correspond, in a measure, to the weathered coal in the matter of extra draft required and consequent lowering of efficiency.

4. *Spontaneous Combustion.* In a recent bulletin by Parr and Kressman[1] it is shown that the initial cause for the heating of coal in piles is the activity it possesses in the matter of uniting with oxygen at ordinary temperatures. The oxidation of pyritic sulphur is only secondary and of a contributory character. These conclusions are also arrived at by Porter and Ovitz[2].

5. *The Storage of Coal.* The ability to store coal in large masses is directly dependent upon an understanding of and properly meeting the peculiar properties of the resinic bodies. This follows as a direct corollary to the preceding topic touching the causes for spontaneous combustion. This fact is susceptible of demonstration in so many ways that one illustration only will be given here. In a recent experiment in storage methods, screenings in car lots from the same mine were stored in two lots, alternating the cars so that the same kind of coal went into each pile. In one case the coal was thrown out by shovel upon a pile which was narrow at the top and wider at the base by reason of the loose particles of coal rolling down the side. As the coal increased in amount, the width of the mass at the top was increased and the material at the base was further augmented by the loose particles which would roll down the side. In this manner, it will readily be seen, a mass of coal was built up having the finer stuff on top, while the base, as it was gradually formed of the loose nut sizes, was open and porous. It may be noted here that this pile was at once in distress. The heating began along the base of the pile and quickly reached the glowing stage.

The other part of this same coal was delivered by belt machinery through a single spout and from that point was moved in large masses by a road scraper and horses and distributed between retaining walls. Of course, these conditions were favorable for very thoroughly packing the mass. This was further assisted by the fact that the coal came in as a rule in a wet condition. This pile of over 2000 tons attained a height on the average of about 12 feet. Numerous temperature pipes were inserted about the mass so that readings could be taken at any desired depth. This pile remained in perfect condition for some months. Recently, however, a discharge of fresh coal from the spout was allowed to form in a conical pile with the loose nut sizes rattling down to the base of the pile. Heating at once began at that piont. These experiments in storage are being continued but they have already gone far enough to demonstrate the fact that coal storage, when it is accomplished, will depend upon a full understanding and explicit following of the principles that govern the behavior of the resinic bodies.

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[1] "The Spontaneous Combustion of Coal." Bulletin No. 46, University of Illinois Engineering Experiment Station, 1910.

[2] "The Spontaneous Combustion of Coal." Technical Paper No. 16, United States Bureau of Mines, 1912.

6. *The Coking of Coal.* It was said at the outset that the resinic bodies of coal had a rather definite melting point and that this was below the point at which decomposition takes place. In a recent bulletin,[1] wherein was given a brief discussion of the theory of the coking of coal, the proposition is offered to the effect that coke formation was dependent upon the presence of a luting body having a definite melting point which was also below the point of decomposition. As an illustration an artificial or synthetic coal was prepared in which glucose played the part of the body with a low melting point and coke breeze served as the bulk of inert non-coking material. The resulting product after passing through the coking process was dense and strong and a good specimen of coke. Now this is exactly the *role* played by the resinic bodies in bituminous coal. The trouble with most coals of the Illinois type is that they have too large a quantity of this resinic material. If we try to coke pure sugar, for example, it blows itself into a light friable mass. If we coke Illinois coals alone we obtain a light porous soft coke. But, like sugar, if we mix in a proper amount of inert material which also will permit of the escape of the gases without blowing the mass into a sort of foaming texture, we shall

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[1] Studies on the Coking of Coal at Low Temperatures. Bulletin No. 60, University of Illinois Engineering Experiment Station. By S. W. Parr and H. L. Olin, 1912.

be able to produce a dense hard coke which bears little resemblance in any of its physical properties to the coke obtained under natural conditions.

Figure A is a good illustration of what may be obtained by mixing an Illinois coal from Vermillion county with coke breeze in the ratio of 1 to 1.

A further illustration touching this function of the resinic material is of interest. By extraction with phenol we are able to divide the coal into the two general types of bodies, the cellulose residuum and the resinic substances. Such a separation is shown in Figures B and C. In B is illustrated the insoluble residue which may be considered as the degradation product resulting from the decomposition of the original cellulose material. The substance C is granular, glassy and rather brittle with a melting point somewhat above 200° and represents the final product of what in the original material was resinous in type. Now if we submit the cellulose body to the coking process we obtain the result shown in B^1. It is a fine powder just as it was before treatment, having shown no tendency whatever towards coking. If now we mix B and C in a proportion corresponding to the original ratio of the two substances, that is, approximately 68 per cent of B to 32 per cent of C, and apply the coking process, we shall have results as shown in C^1.

This latter substance is a hard coke. It should be noted in passing that this property of melting is completely lost if the coal is exposed to the air in a finely divided state or under other conditions as to time, etc., to such an extent that the resinic bodies have become saturated with oxygen. In the case of lignites it is doubtless true that the absence of resinic bodies, or the oxidation to a large extent of such as they may have possessed, is the explanation for the lack of coking properties in such material. Further studies along this line are being continued and it is evident that if ever a serviceable coke is made from Illinois coals it will depend in no small degree upon an understanding of the properties and function of the resinic bodies.

7. *Classification.* It is not intended in this discussion to take up the subject of classification. Enough has been said, however, it would seem, to indicate the important bearing that the resinic

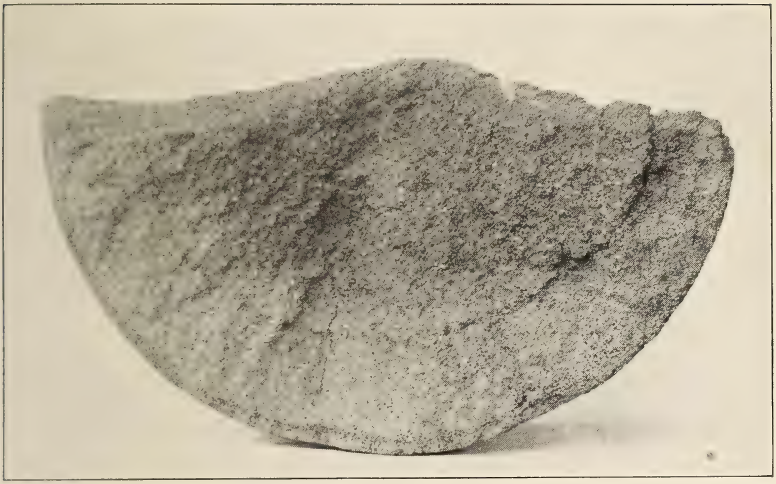

Figure A.

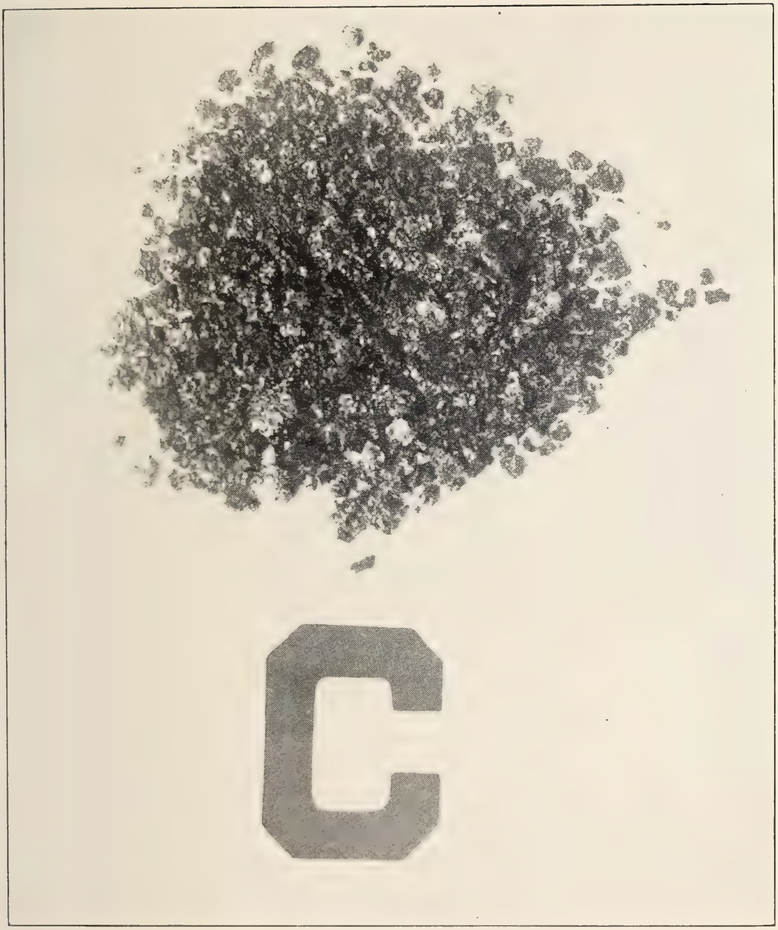

Figure B.

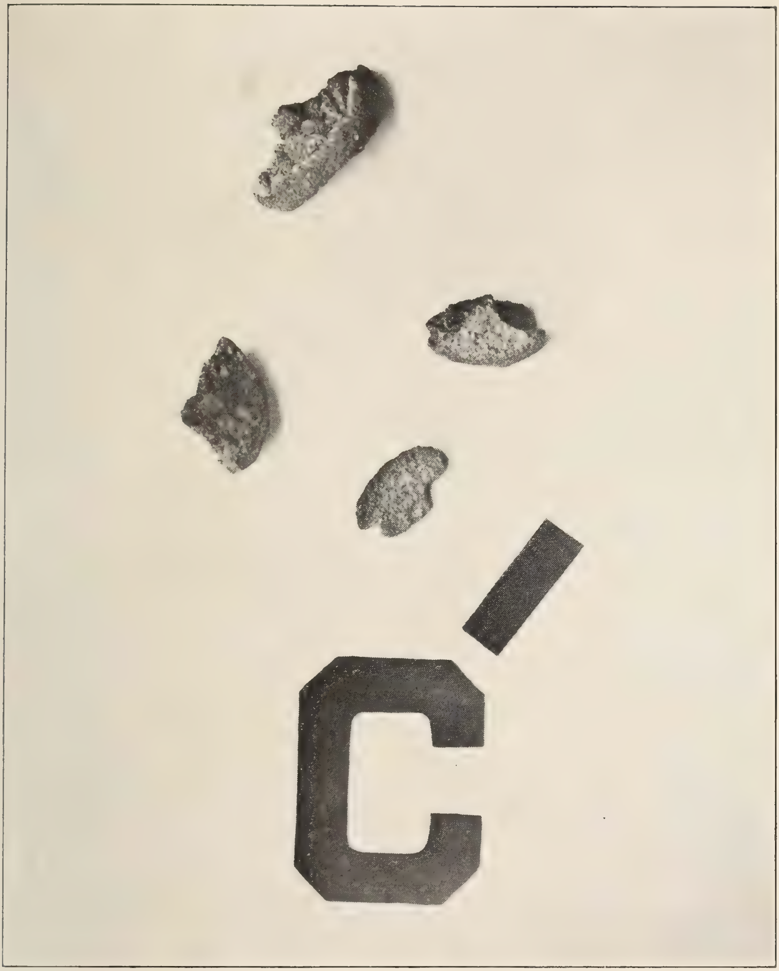

Figure C.

Figure D.

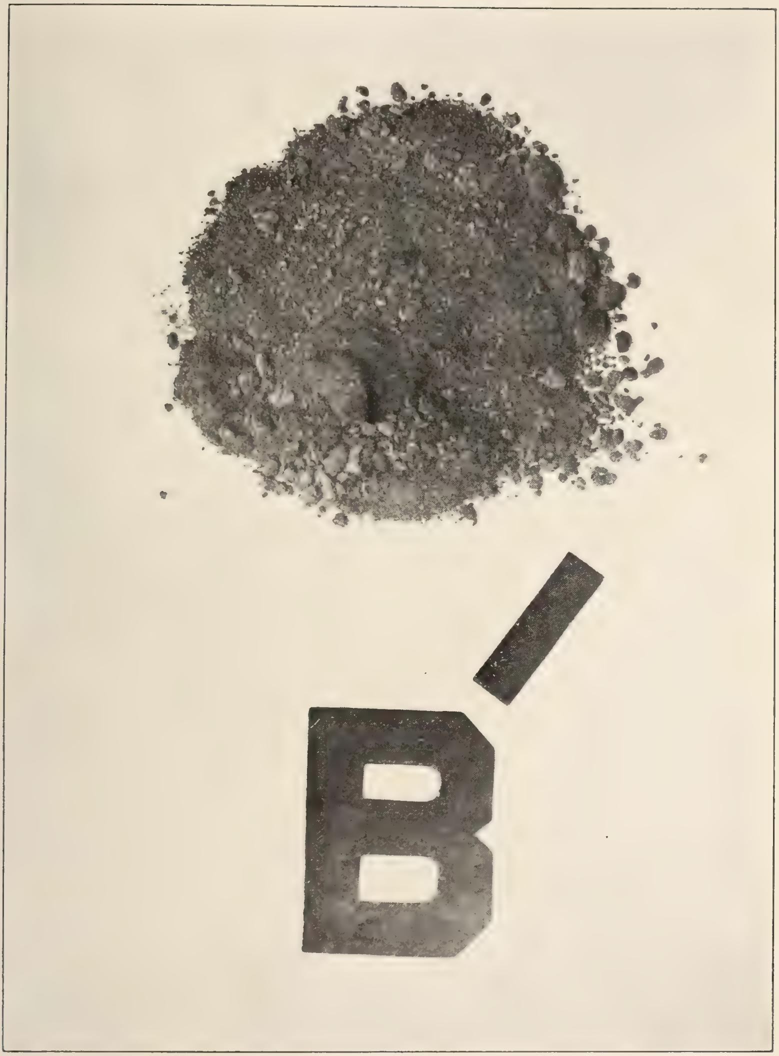

Figure E.

bodies have upon the character of the coal. Evidently, therefore, a more intelligent classification would result only from an extended knowledge of the amount, composition, properties, etc., of this most important ingredient of coal. It is to be hoped that improved analytical methods will bring to our aid much data along this line which at the present time is conspicuously and seriously lacking.

TAR DISTILLATION IN THE UNITED STATES GENERAL DEVELOPMENT AND RECENT PROGRESS

By R. P. Perry
New York, N. Y.

1. Introduction

One feature of the general conservation movement which has attracted much attention in recent years is the estimated use and available supply of coal. For years our household and factory fires have been burning cheerfully,—although in many cases rather smokily,—and we have had an optimistic feeling that our coal mines were inexhaustible. Even now fuel is so plentiful and comparatively cheap in many parts of the country that a large portion of the public forgets that there may be a cold tomorrow. If we travel by night through the Connellsville section we see the glare from thousands of beehive ovens light up the sky and throw picturesque shadows on the hills, but if we study the careful and interesting reports of the United States Geological Survey, we may well be amazed at the apparently reckless annual waste of millions of dollars' worth of by-products,—gas, ammonia and tar,—from these ovens, when converting bituminous coal into coke for the great iron and steel industry. At the same time, those who are familiar with the conditions abroad wonder why they see there so many orderly piles of coal briquettes utilizing the coal dust or slack with a coal tar pitch binder, while in certain regions here they see abandoned culm banks. The explanation is as indicated above,—the past and present generation living for themselves on the easily obtained abundance of our mineral resources, and, if they stop to consider the future, assuming it will take care of itself. Some caustic criticism has been directed at American wastefulness, and it is by no means undeserved, but our foreign friends, who, having more slender coal supplies, raise their hands in righteous horror at our methods, should make due allowance and remember that in some directions the American traveler is equally amazed at the evidence he sees abroad of what appears to him a waste of human labor or of special materials. I can remember, for example, seeing in Germany some miles of solid fence along the railroad made of abandoned steel crossties, one end buried in the ground, set closely touching one another. We would hardly understand such fence construction from our experience and point of view. Similarly, the American manufacturer, accustomed to labor-saving devices at home, is almost speechless when he sees the army of porters cumbrously transporting luggage from boat to dock and *vice versa* at the European ports.

In each case there is sure to be some reason, although not always an adequate one, why conditions are as they are. Some little emphasis is laid on this point as it rises again and again in considering the development of the tar industry in the United States, as it inevitably must be considered in comparison with the development abroad, particularly in Germany. For example, the more scientifically interested traveler who has read of the wonderful coal tar dyes, colors, medicines, etc., and may have seen some of the very large factories in Germany, wonders why such products are not made in the United States. If satisfied with the first tabloid answer available, he will recall what the popular scientific magazines have written about the German superiority in chemical lines, and assume that tells the whole story. The brilliant reputation of our good friends, the Germans, is certainly very well deserved, but numerous domestic and international trade conditions have an equal or more important bearing on why the development of the industry has been along radically different lines here and abroad.

Going back to the initial fact that many million barrels of tar literally go up in smoke from our beehive coke ovens, we find there was in the past a well-defined fear of undertaking by-product ovens for several reasons. In the first place, it took much time to overcome the incorrect feeling that for some intangible reason metallurgical coke was better when made in beehive ovens. Also it was feared there would not be a market for the tar product which would be sufficiently large and remunerative.

Both of these fears have now been overcome, as the successful use of by-product coke has become a fact in the most progressive furnaces, and in general the tar industry has taken up the greatly increased production.

Successful working up of the tar commercially here depends upon finding a profitable use for its main constituent, pitch, as there has been a much greater demand for the oils and the products derived from them than our domestic supply. The foreign use of pitch is mainly as a binder for coal dust or slack in making briquettes. There has recently been a very appreciable development of briquetting in certain sections here, but a curious argument in a circle is still heard at times. On the one hand, some coke operators have said there was no market for additional tar because there was no briquette industry and at the same time briquetting interests have regretted there was no assurance of an adequate supply of pitch binder because the limited output of tar distillation went to make pitches for other purposes which would bear a higher price. Between the two stood the tar distillers, ready and anxious for additional business, but as usual there were other economic conditions having a most important influence.

In many places bituminous lump coal is still so comparatively cheap and abundant that briquettes could not compete and the market for domestic briquettes from Anthracite dust is also somewhat limited. In some cases the geographical location of tar production and possible pitch consumption are so far apart that railroad freights are an insuperable barrier.

Another present and potentially very large use for tar products is in making bituminous road binders, as covered in other papers. I will only mention in passing that although this development in the United States is extremely satisfactory, yet there are very large supplies of Petroleum products and residues designed for road work at low prices, whereas these conditions are reversed abroad, particularly in Germany. In fact, the great increase of road tar abroad has tended to divert much tar, leaving a shortage in the briquette pitch field.

As regards dyes, colors, medicines, special chemical products, etc., derived from coal tar, the conditions are peculiar. Most of these are derived from Benzol, Phenol, Cresol, Naphthalene and Anthracene. Figured on the original tar these total only a relatively small percentage. Further, the finer products require numerous intermediate steps, involving more or less labor, as well as extensive chemical treatment with acids and alkalies. The price of labor abroad, including trained chemists, would probably average not more than about half that paid in the United States. The question as to what the smaller wages will purchase relatively in the way of rent, food, clothing, etc., is an interesting economic study, but does not alter the fact that to the manufacturer the labor costs very much less and enables him to produce cheaper in this respect. In many cases, also, he has an advantage in cheaper chemicals, which play a very large part in the cost of the finished products, running from about fifty per cent upwards. Another most important feature is the patent situation. Many of the finer products derived from coal tar are covered by United States patents held directly or indirectly by a small group of German interests. Dr. Bernard Herstein has recently analyzed this situation in an interesting way.[1] Perhaps one most essential point is that existing United States patent law (differing from that of most foreign countries) permits patenting of the product as such as well as any particular means of production, and further, there is no requirement that the product be made or the process used in the United States. In other words, the German group can and does control a large number of United States patents on these products, and having this protection it is under no necessity of employing American labor or capital in manufacturing here, but can ship to this country from Germany or elsewhere, subject only to tariff regulations. The injustice of a similar situation has recently been recognized by Great Britain which adopted a compulsory working act, with the result that many foreign firms promptly built factories in England. The matter is very important in connection with the features mentioned, producing lower manufacturing costs in Germany. It seems probable, however, that with due justice to inventors, a proper revision of the patent laws will be accomplished before long.

One further rational explanation why these finer products have

[1] Address before Society of Chemical Industry, New York, April 19th, 1912.

not been produced here is that generally there have been uses for the crude or semi-refined products here which netted considerably more than the fine products would have secured. Naturally the tar industry has developed along the lines of least resistance, but already a beginning has been made here in the direction of the finer products, and many of these can be available as fast as there is a demand at prices permitting their manufacture under our conditions. Wherever mechanical handling and machinery plays an important part the United States probably has some advantage.

2. SOURCES OF TAR, AND ESTIMATED QUANTITIES

In the discussion so far, I have referred only to the supply of tar from by-product coke ovens, as this is much larger at present than gas works tar and growing much more rapidly. The earlier coke ovens were built near the mines and steel works of Pennsylvania, but they have followed the blast furnaces as these have moved to the large distributing centers,—it having been found better to haul the coal to such points instead of the coke. Such, for example, is the recent very large development near Chicago, there being by-product ovens at Gary, Joliet, South Chicago and Milwaukee. More recently, under peculiarly favorable conditions of supply of raw materials for iron and steel manufacture, some large coke oven plants have been erected near Birmingham, Alabama. There are many others distributed throughout the eastern half of the United States, some of them located where they can supply illuminating gas to large cities, and others use their surplus gas in open hearth furnaces or other parts of the steel plant. As distinct from by-product ovens, the gas works are located always in the cities, the largest producers of gas works tar being in New York, Chicago, Philadelphia, etc. Most of these gas works produce both coal tar and water gas or oil tar. The differences of quality will be considered later. In some sections of the United States the abundant and cheap supply of natural gas has temporarily, at least, made the operation of gas works financially impracticable.

It is somewhat difficult to get complete data of production of tar, especially of the gas works grade, but it seems probable that, including coke oven and both kinds of gas works tar, the 1912 production in the United States may not be very far from 165,000,000 gallons, of which about two-thirds is coke oven tar. In comparison with this, it has been estimated[1] that if the remaining beehive ovens were converted to by-product ovens, there would be an annual increased production of perhaps 375,000,000 gallons. That the development in this direction has been recently very strong is indicated by the fact that within the year ending in September new by-product ovens have begun operations turning out approximately 35,000,000 gallons of tar per annum.

3. QUALITY OF VARIOUS TARS

Depending upon the coal used, and more particularly upon the size and shape of retorts, the travel which the gases make over the hot surfaces, and the temperatures to which the gas is subjected, the coal tars vary within wide limits. In general with the vapors subjected to the higher temperatures usually characteristic of gas works as compared with coke works, there is an increase in specific gravity and free carbon. For example, the tar from many gas works will average about 1.24 specific gravity at 15.5° C. (60° F.) and about 25% to 30% free carbon by weight, whereas from the by-product coke ovens the specific gravity would average more nearly 1.19 and the free carbon would vary about 5% to 18%, the average being less than 10%.

This so-called free carbon represents a finely divided inert material, largely carbon, which is the portion of tar insoluble in Benzol and Toluol. Perhaps a very small part is carried over mechanically in the coking process, but for the most part it is due to the cracking of the hydrocarbons by exposure to heat, and the higher percentage usually found in gas works tars is from this cause. All gas works tar does not contain a high percentage of free carbon, as I found that in one of the largest European gas works the free carbon in the tar was only about 10%, the percentage having been reduced very much from former figures by increase in the charging of the retorts and some other operating changes. There has recently been some development of the use of inclined and vertical gas retorts in the United States, some of which produce tars lower in free carbon than many coke ovens.

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[1] Figured from data in chapter on "Coke" by E. W. Parker, "Mineral Resources of the United States, 1910."

The free carbon test of the various tars is very important because the percentage has a very considerable influence on the pitch product, and, moreover, it is an indication, to a certain extent, of the quality of oils and bitumens forming the balance of the tar. For many years there was very little coal tar produced in this country, except from gas works operating at very high heats, and the tar was extremely viscous, of high specific gravity and often containing from 30% to 40% free carbon. It was very difficult and expensive to distill such tar, because of the large percentage of water which it always contained and the tendency for excessive carbon to coke on the still bottoms and cause them to burn out. Because of this extreme, the lower carbon coke oven tars were welcomed as of relatively better quality, but today they are not necessarily more valuable as they yield pitches which are extremely subject to temperature influence — being very brittle when cold and easily flowing when heated—and therefore unsuitable for many pitch uses in the United States. The present vastly preponderating amount of very low carbon tars in many places is an extreme as unsatisfactory at times to the tar distiller as was the very high carbon gas tar. Much of the skill of the successful distiller is directed to a very careful study of the various tars he has to handle, the selection of each for the purpose for which it is best adapted, and the judicious combining of various tars to produce physical and chemical properties not possessed by any one of them separately.

The oil tars or water gas tars being largely produced from the petroleum oils used in making and enriching gas represent entirely different physical and chemical characteristics, containing compounds of the olefine series as compared with coal tar containing compounds of the aromatic series. In general they are much thinner and more oily than the coal tars, the specific gravity averaging about 1.07 at 15.5° C. (60° F.) and the free carbon about 1%. The pitches produced from these tars, as might be expected, show, to a much higher degree, the properties of brittleness and susceptibility to temperature changes, and the oils have not given the very great value for timber preservation which is characteristic of the coal tar oils. For these reasons the water gas tars have been much less valuable commercially than coal tars and a large amount has been used as fuel. It is to be hoped and expected that further study will find special fields for the use of water gas tar and its products.

There are some small amounts of various other tars made, such as Producer Gas Tar, Lignite Tar, Wood Tar, etc., but as yet these are not commercially significant, although some of them may develop considerably in the near future.

4. NUMBER AND LOCATION OF DISTILLING PLANTS

There are probably not more than about 40 tar distilling plants in the United States, most of them being naturally located near the producing sources in the section east of the Mississippi and north of the Ohio rivers. The tars will average to weigh about 9½ pounds per gallon and the freights from the gas or coke works are often 25% to 40% of the value of the tar at the shipping point, and for this reason there are many small distilling plants designed to take care of the output from only one or two local sources. At the same time there are large distilling plants in such cities as New York, Chicago, Philadelphia, Boston, Cleveland, Pittsburgh, St. Louis, etc.

5. TRANSPORTATION

Most of the tar in the United States is handled in steel tank cars of about 10,000 gallons' (45 to 50 tons') capacity, such cars being largely owned by the distilling companies because of the need of specially designed equipment which the railroad companies do not furnish. These tank cars are of the usual horizontal cylindrical type, about 7 to 8 ft. in diameter and 28 to 30 ft. long, equipped with a dome on top and outlet valve in the bottom. As tar becomes very thick when cold, it is necessary to have an effective system of steam pipes for heating when unloading. The breakage of these coils, — due largely to severe shocks in transportation—is a very troublesome and expensive experience. Where conditions permit water transportation is used in harbor and sea-going barges from 25,000 to 300,000 gallons' capacity or more. These boats are divided like oil boats into various compartments and equipped with steam coil systems and powerful pumps. In some cases the tar is handled through pipe lines from the producing plant to the distilling works, but long pipe lines, such as are used in handling petroleum oils, cannot be used with coal tar on account of the much higher viscosity of tar, especially when cold.

6. RECEIPT AT DISTILLING PLANTS

From the tank cars or boats, the tar is pumped (after being heated so as to render it more fluid) into storage tanks of varying sizes, generally from 250,000 to 2,000,000 gallons capacity. The large tar plants require many of these tanks so as to keep the various grades of tar separate, there being often four or five main grades and other special ones from time to time. These storage tanks are generally of steel bottom and sides, the roofs and roof supports being of wooden construction as steel roofs are subject to rapid corrosion. The wooden roofs are covered with built-up roofing of felt, pitch and slag or gravel. These tanks sometimes have steam coils covering the entire bottom, but more often only a small section around the outlet suction to permit warming the tar as rapidly as required to supply the pump. The tanks have small outlet cocks on the side distributed from the top to the bottom, so as to permit drawing off as far as practicable any water which was separated from the tar.

7. USES OF CRUDE TAR

Just as there existed a preference in favor of beehive coke, so for many years crude tar as received from the gas or coke works was preferred for some purposes, but a rational study has shown that the water and ammoniacal liquor contained in the tar are detrimental and only a small amount of tar is now being used in the crude state, except in such cases as the tar is used for fuel.

8. DEHYDRATION

It has long been recognized that one of the most troublesome and expensive steps in tar distillation is the removal of the water which is intimately mixed with the tar as received from the gas or coke works. The gas works tar is more troublesome because it contains a higher percentage of water, and, at the same time the water is more difficult to remove because the tar is of greater viscosity and contains more free carbon. Many of the coke oven tars will average between 3% and 6% water, and the gas works tars between 4% and 10% water, although both kinds sometimes contain very much more, up to 40%.

Two main facts to bear in mind explaining why the presence of water is so troublesome are: First, water at 100° C. (212° F.) when changing into steam increases in volume 1646 times, or in other words, a cubic inch of water becomes nearly a cubic foot of steam.

Second, water has the highest known specific heat. Dry tar has a specific heat of about 0.3 to 0.4 depending on the temperature and specific gravity. It will be noted, therefore, that it requires about three times as much heat to raise one pound of water one degree as it does one pound of dry tar.

Many methods have been used to dehydrate tar; perhaps the oldest one being that of allowing it to settle in storage tanks and depending on its lighter specific gravity to cause the water to rise to the surface. This process is aided by heating, intermittent heating with periods of settling being better than continuous heating. Steaming in large storage tanks is, however, very expensive on account of the great loss by radiation, and at best only separates out a portion of the water, particularly with gas works tar. Other methods have been tried, such as centrifugal machines, squeezing the tar between heated rolls or in tanks under pressure, etc., but have not been commercially successful. The plan most in favor is that of heating the tar continuously in thin films under partial vacuum. This method ordinarily reduces the water to less than one half of 1%. Special dehydrator apparatus is in use at some of the large works having a capacity of about 50,000 gallons in twenty-four hours.

9. STILLS

After the tar has been dehydrated it is pumped to intermediate insulated storage tanks and thence as required to the stills for making the numerous grades of prepared or refined tars, soft, medium or hard pitches. Contrary to the German and English practice of using vertical stills with concave or convex bottoms, the still development in the United States has been mainly along the lines of horizontal cylinders. Quite a number of vertical stills have been used for particular conditions and have certain valuable characteristics. Many of the German stills up to recently held from 20 to 30 tons of tar, but the usual stills in the United States hold twice as much or more. One fact which is said to be largely responsible for the smaller unit abroad has been their desire to avoid night operation, whereas it is quite common with us. It seems quite probable to me that before very long we shall have in our large works in the United States three eight hour shifts, with continuous running of the stills day and night, certain other portions of the works, however, operating only in the daytime.

Different types of brick still settings have been used with the general purpose of avoiding over-heating and burning out the still bottoms and at the same time getting the maximum heat efficiency from the coal, liquid or gas fuel. Depending upon the local conditions and the quality of the oils and pitch desired, the stills are run with mechanical, steam or air agitation, or vacuum in addition to the usual heating. The temperature reached in the liquid in the still depends on the grade of pitch desired, the higher temperatures causing more oil to be distilled off and leaving a harder pitch, but the highest temperature does not usually exceed 400° C. (752° F.). The operation of these stills is intermittent, consisting of charging, distilling off the desired amount of oil, removing the residue by gravity, blowing or pumping, and repeating. Continuous stills have certain theoretical advantages, but have not been adopted in practice in the United States. Successful continuous distillation requires a uniform grade of tar and a demand for a very large amount of one grade of pitch for uses which permit quite a little latitude in range of melting point. Several continuous processes have been tried abroad, but have not met with general success, although I know of one very large plant where one continuous process is reported to have been operated with good satisfaction over several years at low cost. In many cases there has been too much fluctuation in the test of the pitch and oil product, but I feel sure the continuous process will come under conditions likely to develop in the future in this country.

10. OILS, CONDENSATION, USES, ETC.

The vapors escape from the still through a large vapor pipe to the condenser coils. All of the vapor pipe and preferably all of the coils should be made of flanged cast iron pipe using thin asbestos gaskets. In any case, it is necessary to use cast iron pipe as far as one or two turns below the water in the condenser tank, for the reason that certain of the vapors, particularly some of the ammonia compounds in the vapor form, attack wrought iron or steel very strongly, but have comparatively little action in the condensed liquid form. This is one reason why the tops of the stills sometimes eat through before the bottom. Most of the tars at present contain much less ammonia compounds than heretofore, as the ammonia is for obvious reasons recovered at the gas works or coke works.

The condenser tanks are usually square or oblong, with similar shaped coils, the pipes ranging from six inches down to three inches. The length of the coil depends on the size of the still, the temperature of the available water, etc. The condensed oils are collected and measured in small steel tanks of about 100 to 400 gallons, capacity, depending on the size of the stills and other conditions. These small tanks are provided with sight glasses and empty by gravity into a manifold pipe system leading to various large receiving tanks. From three to five fractions are usually collected, such as Light Oil, Carbolic Oil, Middle Oil, Heavy Oil and Anthracene Oil. The percentage of each fraction varies with the tars used and the commercial requirement for the oils, and no exact figures can be given. The total per cent of oil secured in making pitch is generally from 20% to 50% by weight. From the receiving tanks the oils are pumped to large storage tanks for shipping or further refining.

The Light Oil, that having a specific gravity lighter than water, is separated from the water and then used as crude solvent in paints or for cleaning greasy gears, etc., or it is refined to Benzol, Toluol, Solvent Naphtha, etc.

The carbolic fraction is usually collected so as to give the maximum yield of tar acids consistent with getting at the same time as liquid an oil as practicable. The carbolic oils, crude or redistilled, are cooled by natural or artificial refrigeration to remove excessive naphthalene and sold as crude Carbolic Acid for disinfectant manufacture. Inasmuch as from most tars the carbolic oils seldom contain more than 10% to 15% tar acids, and the usual standard required is 20% to 25%, it is necessary to add extra acid as required. This is obtained by extracting the acids from other carbolic oils, or the middle and heavy oil fraction. The extraction process is a simple one of treating the oil under suitable temperature conditions with caustic soda solution, separating the Sodium Carbolate and throwing the acids out with Sulphuric Acid or more commonly with Carbonic Acid Gas. The extracted acid is then redistilled and separated into Carbolic Acid and Cresylic Acid. The former is the basis for the purified Crystal Acid.

The demand for disinfectants is increasing with the campaign to reduce infectious growth and many excellent grades have been made for years, but at the same time many grades have been made unskilfully and are of no real value beyond the psychological one of having a good healthy odor. Just as a standard of purity or strength is required for foods, drugs, etc., it is reasonable to have a standard of efficiency for disinfectants, and the standard most widely recognized is the Rideal-Walker method of comparing with pure Phenol under special conditions.

The large use for the Middle, Creosote and Anthracene oils is for wood preserving, and they are fractioned so as to meet the various specifications of the wood preservers. This industry is a very large one and growing rapidly, as the conservationists have pointed out to large users of timber the definite fact that the local supply is gone and the more distant forests are being rapidly depleted. The railroads were among the first to appreciate the situation. Many of the large systems have had timber preserving plants for years and now very few are without one or more of such plants. Other papers cover this creosoting industry, but in this connection it is of interest to note that the supply of creosote oil in the United States is much less than the demand and the importations largely from England and Germany during the year 1911 were about 45,000,000 gallons.[1] This quantity represents, perhaps, about two-thirds of the total consumption.

[1] House of Representatives Report No. 326, page 65.

Recently there has been a very large increase in the use of creosoted wood blocks for paving as they make an ideal pavement under many conditions, particularly where a quiet pavement is desired in the business sections of large cities. Also there is an increasing demand for the higher distilling grades of creosote oil suitable for surface treatment of wood (preferably by the hot and cold tank method), and high grade domestic oils are now produced in large amounts.

Another use of creosote oils is for shingle stains, either plain or with colors. The use of certain fractions for Diesel and similar internal combustion engines in Germany has not developed here as yet because there are other more remunerative uses and in this special field there are various cheap petroleum products.

11. REFINED TARS AND PITCHES

Depending upon the amount of oil distilled off, we have as the residue in the stills either thick viscous tar or soft, medium or hard pitch. Sometimes the point for stopping distillation is determined by the still temperature or by the amount and test of the oil distillate, but such is possible only with a uniform grade of tar, and more generally samples are drawn from the still itself and tested, the distillation being continued until the desired grade is secured. The usual test is for melting point, which is determined by various arbitrary tests, as there is no such thing as a true melting point of a pitch. A general classification according to the half inch cube melting point method would be as follows:

Distilled Tars having a melting point less than 32° C. (90° F.)

Soft Pitch having a melting point from 32° C. (90° F.) to 49° C. (120° F.).

Medium Pitch having a melting point from 49° C. (120° F.) to 71° C. (160° F.).

Hard Pitch having a melting point from 71° C. (160° F.) to 99° C. (210° F.).

Very Hard Pitch having a melting point from 99°C. (210°F.) upwards.

When the right grade has been secured the distillation is stopped and the still emptied either by blowing the contents direct from it by means of dry steam, or by drawing the contents by gravity to a special tank from which it is blown as before to storage tanks or coolers. From these the refined tar or pitch is later loaded into tank cars or tank wagons, or run into barrels, iron moulds or pitch bays, depending upon conditions.

Various grades of refined or prepared tars are made for road binders, for mixing tar concrete used under wooden floors in factories, for painting felt roofs and for saturating dry felt in making the various kinds of roofing paper. The grades are usually determined by specific gravity, distillation, free carbon, viscosity, penetration, evaporation, etc.

The soft pitches are used for road binders, and alone or in combination with tarred felts for waterproofing the foundations of buildings, etc. In addition to the tests above, the so-called melting point is noted.

The medium pitches are very largely used for constructing "Tar and Gravel "—more properly "Pitch and Felt" roofs used on nearly all large factories having comparatively flat roofs. The construction consists of applying alternate layers of tarred felt and coal tar pitch, the laps or joints being broken and the top layer of pitch having imbedded in it clean gravel or crushed slag. The standard construction to-day calls for at least five layers of felt besides the usual layers of pitch and gravel or slag. Other large and growing uses are that of paving pitch for filling the joints of brick, stone and granite pavements, and for waterproofing bridges, etc. These grades of pitch are usually shipped in barrels.

The hard pitches are most largely used for briquetting coal dust or slack. This promises to become a very large industry in the United States later on when lump coal is more expensive, and has already become well established in certain sections where briquettes are made from anthracite dust for domestic use.

The very hard pitches are used for electric carbons and batteries, for insulating, for making "clay pigeons" or "flying blackbirds" used for target shooting, for making core compound used in foundries. These pitches are so hard that they can be ground to powder. The hard pitches are run hot either into iron moulds or into pitch bays or pits and there allowed to cool. The iron moulds can be emptied when cold and the pitch can be dug or mined from the bays and is handled to cars for shipment. Most of these hard grades are shipped in bulk so as to save packages and the freight on same.

12. ROOFING PAPER

One important branch of the tar industry is the manufacture of roofing felts, largely a mechanical process and therefore one in which we are further advanced here than abroad. The felt forming the basis of these roofings is made from mixed old woolen and cotton rags, largely cotton, along the lines similar to other paper manufacture. Various widths are used, generally thirty-two inches or thirty-six inches, and the paper varies in thickness from the No. 15 to the No. 80 of the paper maker's scale, weighing from 3.1 to 16.7 pounds per one hundred square ft., single thickness. This felt is made in large rolls weighing about 400 to 600 pounds.

The simplest form of manufacture is the saturation of such felt by passing the sheet continuously through a bath of refined tar, then through revolving squeeze rolls, which remove the surplus tar, and the tarred felt is wound into measured rolls often containing 400 to 500 square ft. This kind of tarred felt is largely used in the "Felt and Pitch" roofs and also for waterproofing and sheathing purposes.

The so-called Prepared or Ready-to-Lay roofings are of many different kinds, one standard form consisting of two or three layers of tarred felt cemented together with pitch and having a plain surface or one covered with pitch or composition into which, while hot, is imbedded crushed Feldspar or small pebbles.

13. REFINED PRODUCTS

The refined products, as noted above, are largely of the general Benzol, Naphthalene, Anthracene group. Much more crude Benzol is now obtained by washing coke oven gas in oil scrubbers than is obtained from coal tar distillation. This gas oil or the light oil from distillation is redistilled, treated with Sulphuric Acid, washed, treated with soda and then distilled with steam. Fractions are collected distilling within certain limits, such as Pure Benzol, 90% Benzol, Toluol, Xylol, etc. These are very largely used in this country as solvents, especially in paint and varnish lines, for making varnish remover and as a basis for Nitro-Benzol, Aniline, etc.

Crude Naphthalene is purified by crystallizing, passing through centrifugals to remove the oily portions, distilling, treating with Sulphuric Acid and crystallizing or subliming.

Large amounts are used for moth balls. Specially purified Naphthalene is the raw material for making many coal tar colors, this line being hardly touched in the United States, for reasons noted above. Recently some new wax products have been introduced having peculiar dielectric properties, and promise to develop an important field. These are made from Naphthalene by a chlorination process.

Anthracene as the basis for Alizarine was of great importance at one time, but is not largely purified at present because of the low market price.

Although brilliant chemists have been studying tar products for years, yet the number of constituents and the innumerable combinations which can be produced by treating them with other compounds leaves plenty of opportunity for the future. The recent opening up of various pressure, electrolytic and catalytic processes will probably give us more astonishing products from tar than any yet secured, each success stimulating the world of scientists to further efforts. That the United States will ultimately take a prominent position in the industry is to be expected from the future great supply of tar in this country, and the energy of her students who admire the success of the German, English, French and Swiss chemists and are determined, if possible, to go one or more steps further. Fortunately there is ample room for all in this fascinating line of work.

THE SPONTANEOUS COMBUSTION OF COAL

By Horace C. Porter and F. K. Ovitz
Bureau of Mines, Pittsburgh, Pa.

Fires are frequently caused in coal piles, coal bins and coal bunkers of ships by the accumulation of heat from so-called spontaneous chemical reactions. Spontaneous heating is responsible for large losses in stored fuel. Even though actual fire does not occur, the deterioration by oxidation rapidly increases as the coal becomes heated. The storage of large quantities of coal, 500,000 tons and more, at one point for a period of one year and longer, is becoming a common practice, by reason of the uncertainties of supply due to strikes and transportation difficulties; therefore the reduction of losses to a minimum in such valuable quantities of stored material is an economic problem of importance.

That the self-initiated action of the air upon coal at the ordinary temperatures is sufficient in amount and in rate to cause, under suitable physical conditions, a continuous rise of temperature to the burning point, most authorities are agreed. An opinion, however, is quite commonly held by consumers of coal that the coal substance itself is rarely, if ever, responsible for the initiation of such heating, but that the fault lies always in certain impurities, such as iron pyrites, fire-clay, grease, oil or other extraneous matter.

It is the purpose of this paper to show by experimental evidence that the carbonaceous material — the coal substance proper — through its gradual oxidation, is chiefly responsible for the initial development of heat which results in spontaneous combustion; and that certain physical conditions, subject to control, are the chief factors which determine the continuance of the temperature rise to the point of ignition of the coal. It will be shown that there is at ordinary temperatures a reaction of coal substance with oxygen — a peculiar reaction, not setting free an amount of CO_2 equivalent to the oxygen absorbed; that this reaction, when it occurs at a somewhat elevated temperature, is exothermic in character; and that the action of oxygen on the coal substance itself is greater than the action on the sulphur compounds occurring in coal, even when the latter action is promoted by the presence of moisture. It will be shown that at 100° C. coal absorbs oxygen and develops heat while undergoing entirely insufficient sulphur oxidation or carbon oxidation to account for the oxygen absorbed, even though the coal be moistened to promote the sulphur oxidation.

There will be shown further, by laboratory experiments and by tests and observations on actual cases of spontaneous fires in commercial practice, the large importance of physical conditions such as fineness of the coal, rate of air supply to the interior, and bulk of the mass, in determining whether the slowly developed heat shall accumulate or be entirely dissipated.

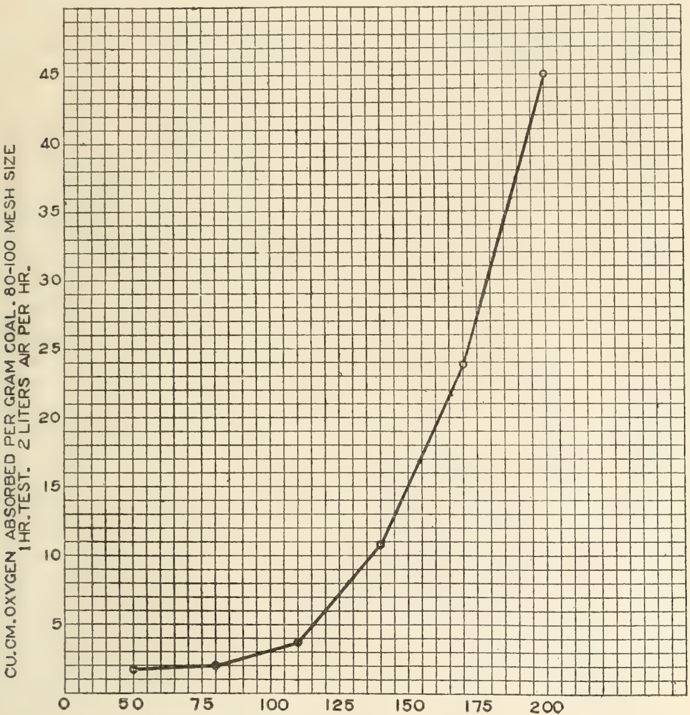
First there is to be considered the cause of the initial development of heat. It has been proven beyond any doubt[1] that freshly mined coal absorbs oxygen rapidly at ordinary temperatures without a corresponding formation of CO_2. The rate of this action diminishes rapidly at first, but after a brief period becomes more or less uniform and continues so for a long time. In certain experiments whose primary object was the study of the escape of methane from coal,[2] it has been found that 10 kilograms of a southern Illinois coal, crushed to pass a one-half inch screen, exhausted the oxygen (1140 cc.) from the air of its containing vessel in four days or less from the time it was mined. Similarly, a coal from the Pittsburgh bed (10 kilograms) absorbed 1080 cc. oxygen from the air of the container in one day. These coals, and several others used in similar experiments, continued to absorb oxygen as long as the experiments were conducted (5 to 10 months), at a rate depending on the partial pressure of the oxygen supplied. Carbon dioxide was not produced in quantities greater than 1/10 of the oxygen absorbed, this amount being doubtless partially made up of occluded CO_2 escaping from the pores of the coal. Analyses of the coal showed, also, that there had been a similarly disproportionate oxidation of the sulphur present. Experiments have shown[3] furthermore that coal subjected to a

[1] Boudouard. Comptes Rendus 148, 284, and Bull. Societé Chimique de Paris, 5 (4), 377.

Tech. Paper 2, Bureau of Mines, Escape of Gas from Coal.

[2] Techn. Paper 2, Bureau of Mines. The Escape of Gas from Coal.

[3] Porter and Ovitz. Jour. Amer. Chem. Soc. 30, 1490. Archibald and Lawrence. *Jour. Ind. & Eng. Chem.* 4, 258.

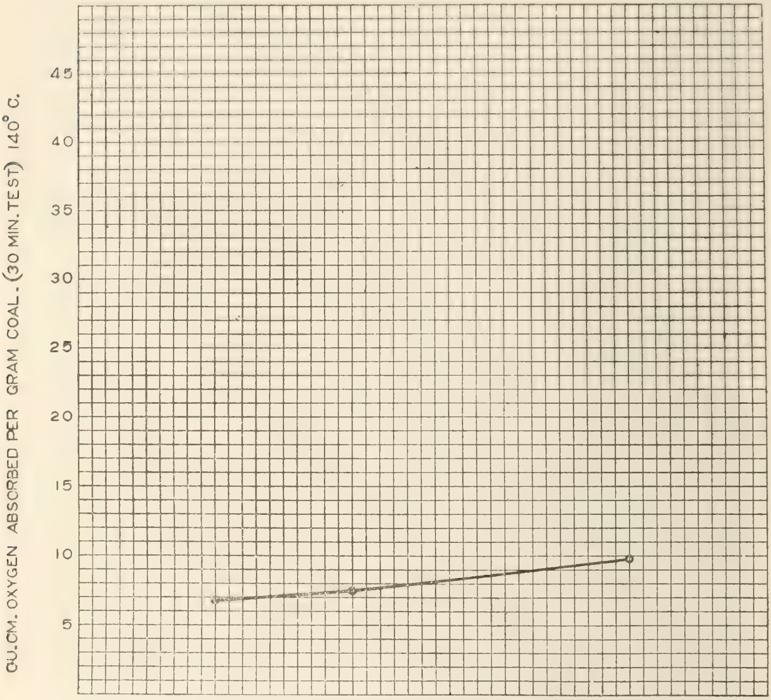


RATE OF OXIDATION OF DRY COAL
EFFECT OF TEMPERATURE

current of dry air at 105° C. for two hours increases in weight 0.4 to 0.5% by absorption of oxygen and produces less than 1/10 that amount of CO_2.

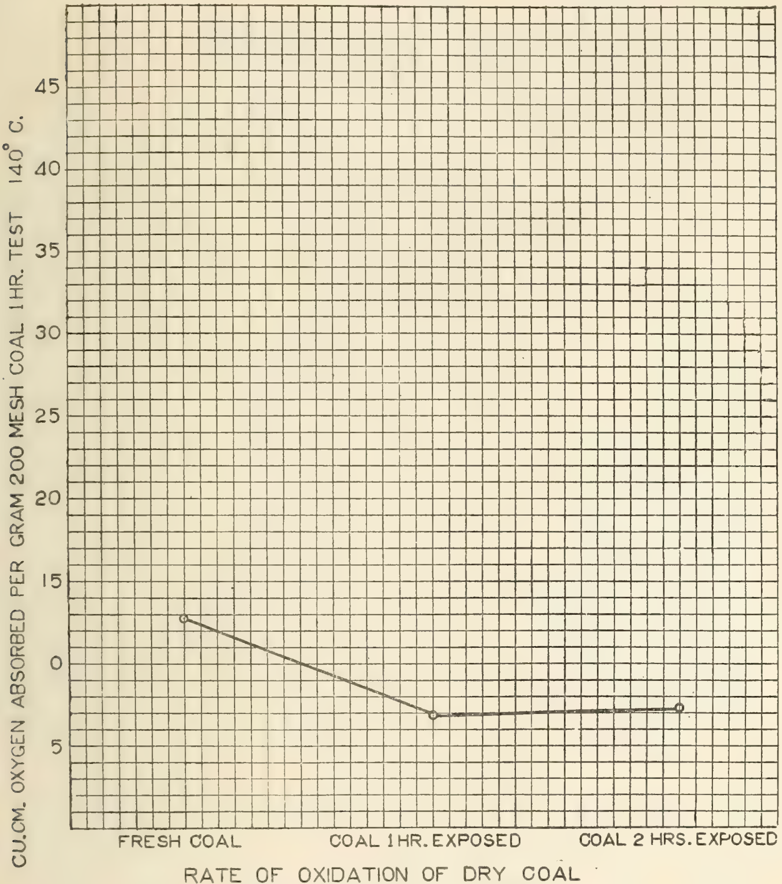
Although the fact is thus well established that this oxygen absorption occurs, there is as yet no experimental data establishing the rate of the production of heat by the reaction at ordinary temperatures. On accelerating the action by conducting it at somewhat elevated temperatures, its exothermic character is clearly shown.

Different coals, powdered to 60 mesh size and dried in a current of CO_2, were heated at 140° C. and 162° C. in a current of 50% oxygen. The coal samples were 15 grams in size, the oxygen



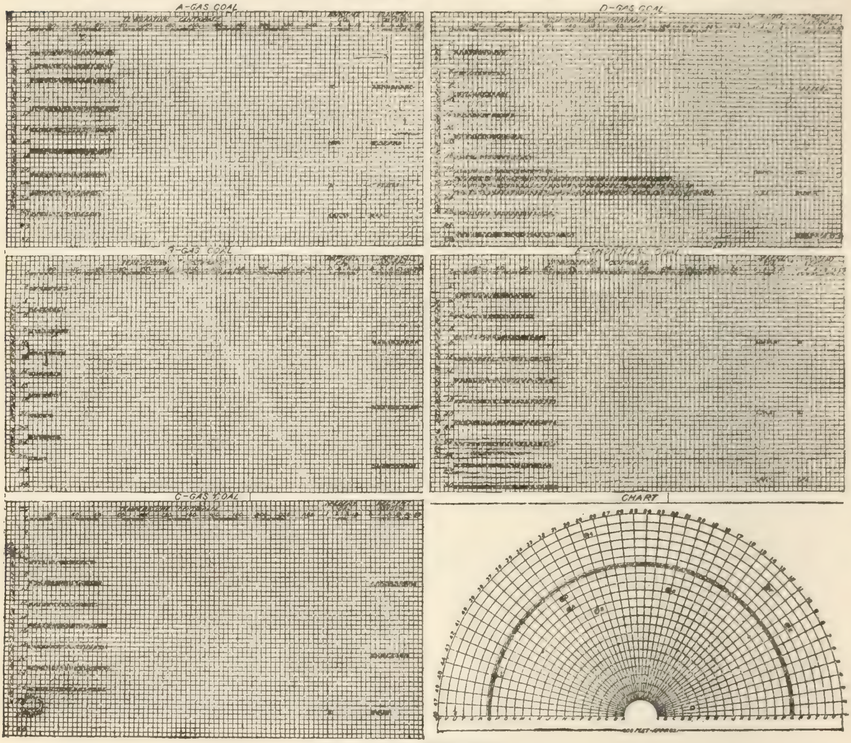
RATE OF OXIDATION OF DRY COAL
EFFECT OF RATE OF AIR SUPPLY

was dried and preheated, and the rate of flow of oxygen was four liters per hour. The rise of temperature in the coal sample at the surface where the oxygen enters it is shown in Figure 1, for several different coals. That sufficient heat was developed under these conditions to cause ignition in several cases is evident from the results. In order to determine the relative CO_2 formation, a test was run on an Illinois coal (No. 48) after drying in nitrogen. The coal ignited in the oxygen current with a bath temperature of 150° C. and there was a production of 2.2 ccs. of CO_2 per gram of coal, equal to about 1/10 of the oxygen absorbed. At the temperature used, therefore, the oxygen consumed goes only in small part to burn carbon to CO_2.



RATE OF OXIDATION OF DRY COAL
EFFECT OF PREVIOUS EXPOSURE

The influence of the size of the coal on the results of this test is shown in Figure 2. Comparative tests were run on 60 mesh samples and on 20–40 mesh samples, the former size showing much more rapid oxidation than the latter. In Figure 3, the influence of the sulphur content of the coal on the result of the test is shown. The Missouri coal (No. 209) containing 3.34% sulphur, was subjected to a floating treatment which separated it into a lighter portion carrying 2.61% sulphur and a heavier portion carrying 5.33%. The heavier portion showed no greater tendency to



TEMPERATURES IN COAL PILES
COMPARED WITH COMPOSITION OF AIR IN PILE

develop heat in the oxidation test. It must be noted, however, that this test was made upon dry coal and that the oxidation of sulphides of iron at the temperature used probably does not take place at all except in the presence of moisture.

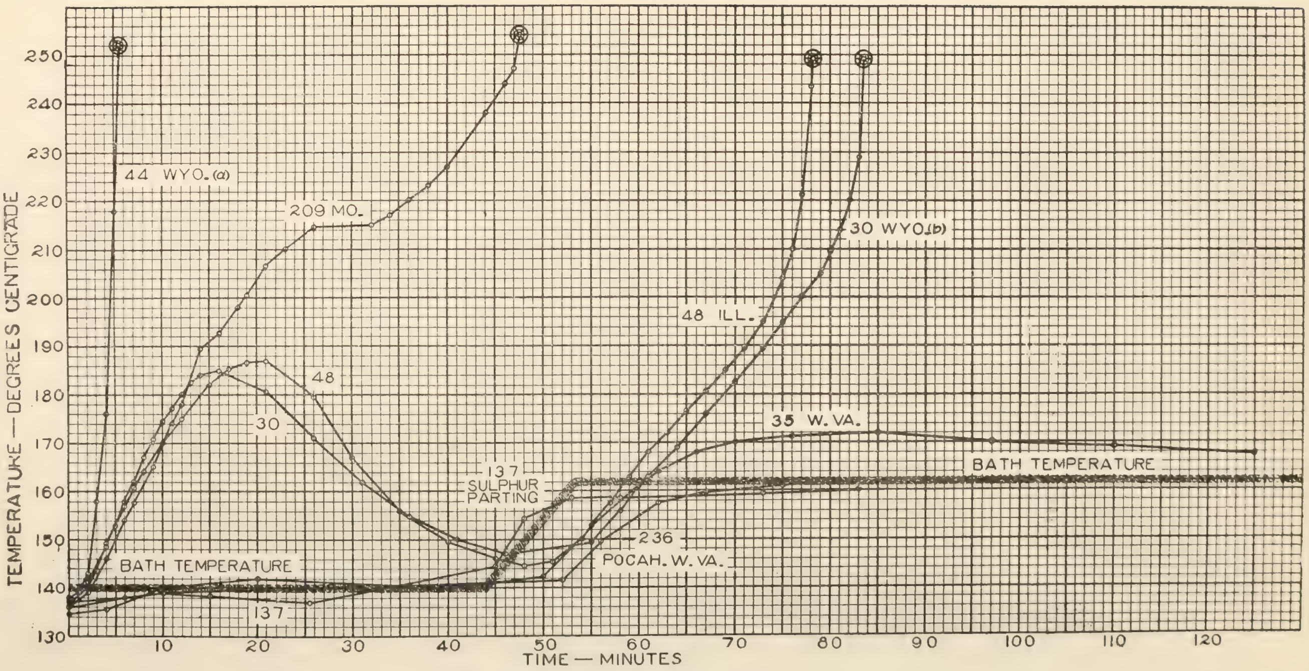
It is evident that at 140°–160° C. there occurs a heat-producing, oxygen-absorbing action of oxygen on coal, varying in intensity with the type of coal and not consisting of the usual combustion of carbon to form CO_2.

Further experiments, supplementing the above, were performed on samples of Illinois coal at the temperature of boiling water. One of the samples was of a coal containing over 4% sulphur, and an endeavor was made to show the relative ease of oxidation of the carbonaceous material and of the sulphur-bearing material at this temperature. After drying 15 grams of 60 mesh coal in a partial vacuum at 98°–99°, a current of 60% oxygen was passed over the coal at the rate of 4 liters per hour. In a parallel experiment the coal was moistened with 15–20% water and kept moist during the test by saturating the oxygen current with moisture at 80°–90°C. The results were as follows:

1. Coal No. 23 (Saline Co., Ill.); total S, 1.52% (on air-dried coal); sulphur in form of sulphate, .018%; sample in ¼″ size had been kept four years in the laboratory, submerged under water. (*a*) Dried coal in current of oxygen gave a rise of temperature of 3.0° above the bath; absorbed 93 cc. of oxygen in one hour; produced 3.9 cc. CO_2; and no increase of sulphur as sulphate. (*b*) Wet coal in a current of oxygen gave an oxidation of .0024 grams sulphur to sulphate, equivalent to the absorption of 3 cc. of oxygen.

2. Coal No. 13451 (Vermilion Co., Ill.); total sulphur 4.38% (on air-dried coal), sulphur as sulphate .098%; sample used had been mined four months and kept (without pulverizing) in a closed can during that period. (*a*) Dried coal, in oxygen current, gave a rise of 1.5° C. absorbed 110 cc. oxygen in one hour, produced 6.2 cc. CO_2 and no increase of oxidized sulphur. (*b*) Wet coal in the oxygen current gave an oxidation of .003 grams sulphur to sulphate, equivalent to the absorption of 4.0 cc. oxygen.

3. Mixture of two parts coal No. 137 (sulphur parting from Sun mine, Fayette Co., W. Va., 33.2 per cent sulphur), and one part coal No. 13,451 (Vermilion Co., Ill.), containing in the moisture approximately 43.3 per cent pyrite and 42.9 per cent carbonaceous material; sulphur as sulphate in the mixture, 0.502 per cent. (*a*) Dried coal, in oxygen current, gave a rise of 1.0°C.; absorbed 69 cc. oxygen in 1 hour, produced 3.5 cc. CO_2, and no increase in oxidized sulphur. (*b*) Wet coal in the oxygen current gave an oxidation of .068 per cent sulphur to sulphate or .0102 gram, equivalent to the absorption of 14 cc. oxygen. In this experiment nearly equal quantities of FeS_2 and organic material were present, and the oxidation of the latter in the dry condition was almost five times that of the pyrites when wet (the pyrites oxidation being practically nothing when dry).



SPONTANEOUS COMBUSTION

BEHAVIOR OF COALS IN CURRENT OF 50 PERCENT OXYGEN AT 140° AND 160° CENTIGRADE

RATE OF FLOW OF OXYGEN—4 LITERS PER HOUR

⊛ =IGNITION

It is shown by these experiments that at 100° C. the attack of oxygen on the carbonaceous material of coal is fully as rapid as it is upon the sulphur-bearing material, weight for weight. As to which action is more rapid, however, at ordinary temperature, these experiments offer no conclusive evidence nor do they show the comparative quantities of heat developed in the two actions.

An experiment reported by Parr and Kressmann[1] shows indirectly a relationship between the quantities of heat developed by oxidation of the coal substance proper and by oxidation of the sulphur. A 40-lb. sample of Illinois coal (1.35% sulphur) crushed and sized between $\frac{1}{8}''$ and $\frac{1}{4}''$ was thoroughly wet down and placed in ovens for successive periods of three days each at 60°, 80° and 115° C. At the end of the heating the coal had dried out and its temperature had risen 21° C. above that of the oven. There was an increase in the percentage of sulphate sulphur from .0189% to .0317%, or .0128%, corresponding to an oxidation of 4.26 grams of FeS_2 with a calculated heat production of 11.4 large calories. The total heat production corresponding to 21° rise of temperature in the coal, disregarding the warming and evaporating of moisture and assuming the specific heat of coal as .25, was $(18.12 \times 0.25) \times 21 = 95$ large calories, more than eight times that due to sulphur oxidation.

The same authors report other tests on coal more finely powdered, in which a much larger amount of sulphur oxidation occurred. There resulted in that case a much greater rise of temperature and, therefore, a larger heat production, which at first glance appears to be in a large measure accounted for by the sulphur oxidation. But, on account of the slow drying of the finely powdered coal, the heat developed was probably utilized to a large extent for evaporating moisture, and its true amount cannot, therefore, be accurately estimated for comparison with the calculated heat of sulphur oxidation. Other tests by the same experimenters indicated a greater heat increment in coal containing 5.0% FeS_2 than that in the same coal with 1.65% FeS_2. On the other hand, coal with 3.0% FeS_2 gave still greater heat production than that with 5.0%. The authors themselves state that in their experiments the thermometer did not always indicate the maximum

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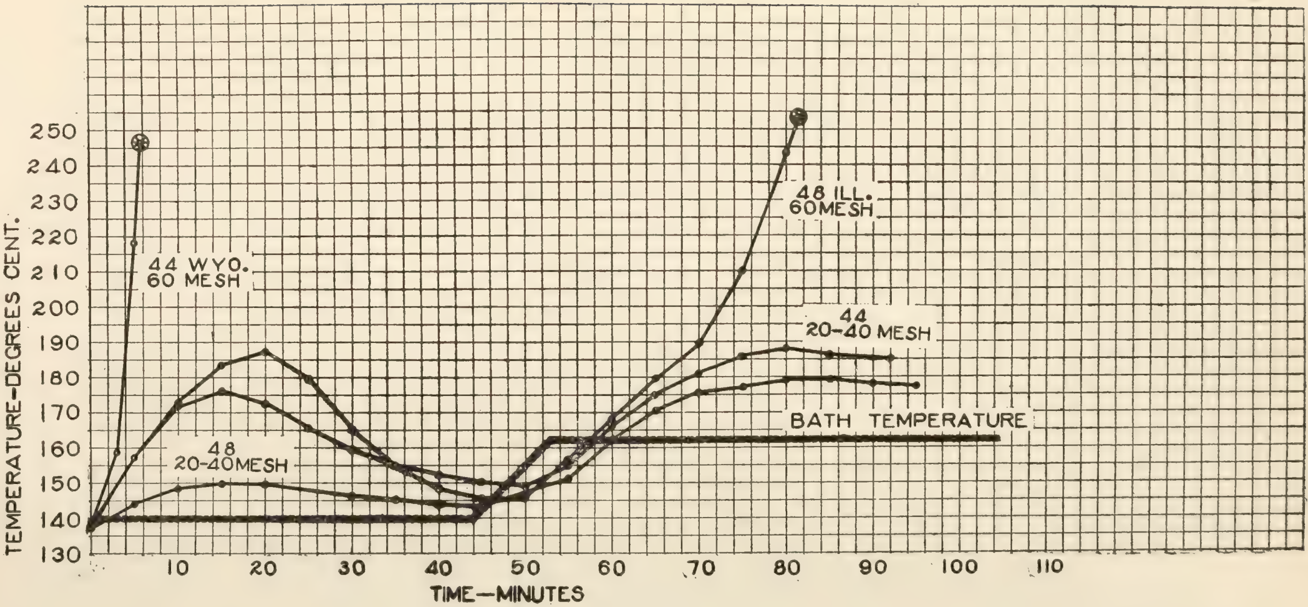
[1] Bull. 46, Univ. of Ill., Eng. Exp't Sta., pages 25 and 31.



SPONTANEOUS COMBUSTION

BEHAVIOR OF COALS IN CURRENT OF 50 PERCENT OXYGEN AT 140° AND 160° CENT.
EFFECT OF SIZE OF COAL
⊛ = IGNITION

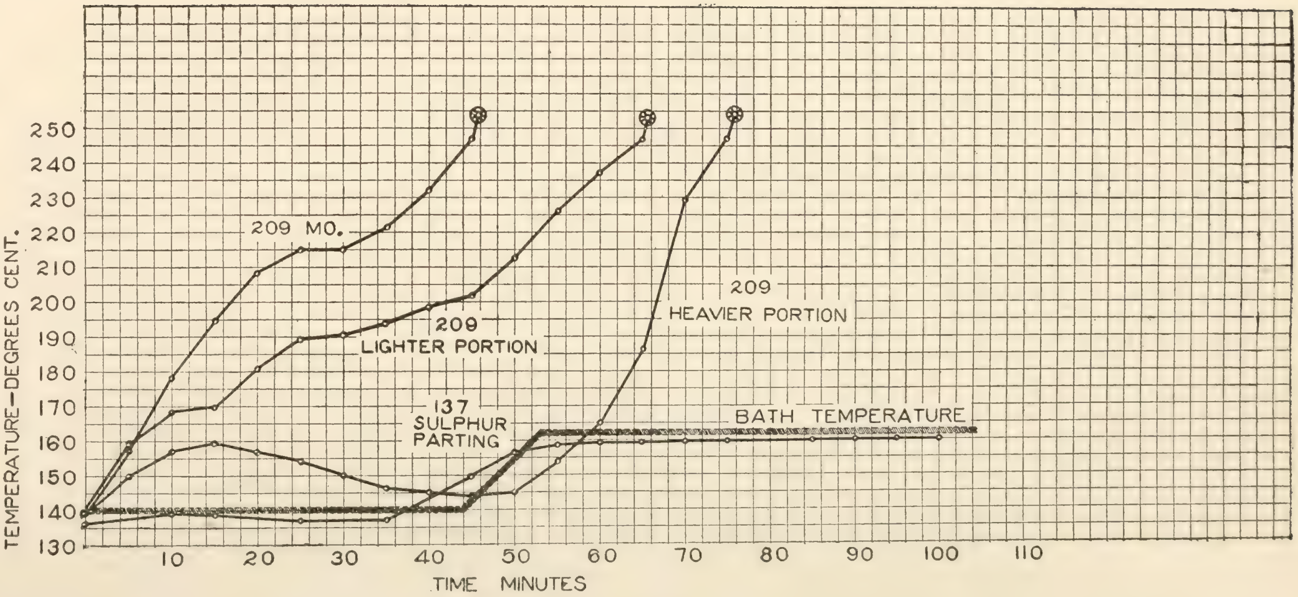
temperature rise in the coal, and that in the tests on wet coal the amount of water varied somewhat, thus affecting the time of drying and the rise of temperature in a given time. It appears, therefore, that the experimental evidence at hand is not sufficient to establish any positive effect of sulphur on spontaneous heating.

Evidence is at hand which shows less oxidation at ordinary temperatures in the iron pyrites (FeS_2) of coal than in the coal substance proper. A sample of 12 kilograms of a Southern Illinois coal (No. 49), under ½″ size and not dried, was kept in a bottle for five months in a slow current of oxygen, about ½ liter of 95% oxygen being admitted to the bottle every day. The coal absorbed during this five months' period about 500 cc. pure oxygen per 100 grams. The moisture content of the coal was 3.86% and the sulphur 1.46%. After standing 2½ years longer in the stoppered bottle, the sulphur present in the oxidized condition was determined and found to be .035%. Assuming that no oxidized sulphur was present in the coal as mined, this is equivalent to an oxygen absorption of about 50 cc. per 100 grams of coal, or one-tenth of the total oxygen absorption which had occurred in the first five months. This amount is probably about double the true figure, since on the basis of analyses of other coals from the same region the coal probably contained about .018% oxidized sulphur when mined.

Somermeier[1] determined the amount of oxidized sulphur in several samples of "high sulphur" coals after standing 1½ years at ordinary temperature in the moist condition as received from the mines. He found in samples kept in closed bottles, a maximum of 0.47% oxidized sulphur and in one sample kept in an open sack 0.37%. This is probably about 1/10 of the amount of total sulphur present (the exact amount of total sulphur, however, not being stated).

A number of samples of weathered coal collected from outdoor coal piles after six months' to one year's storage, and in some cases after spontaneous heating had occurred in the coal, have shown on analysis not over 1/10 of the total sulphur to be in the oxidized condition.

[1] Jour. Amer. Chem. Soc., 26 (1904), p. 764



SPONTANEOUS COMBUSTION

BEHAVIOR OF COALS IN CURRENT OF 50 PERCENT OXYGEN AT 140° AND 160° CENT.)
EFFECT OF SULPHUR IN COAL

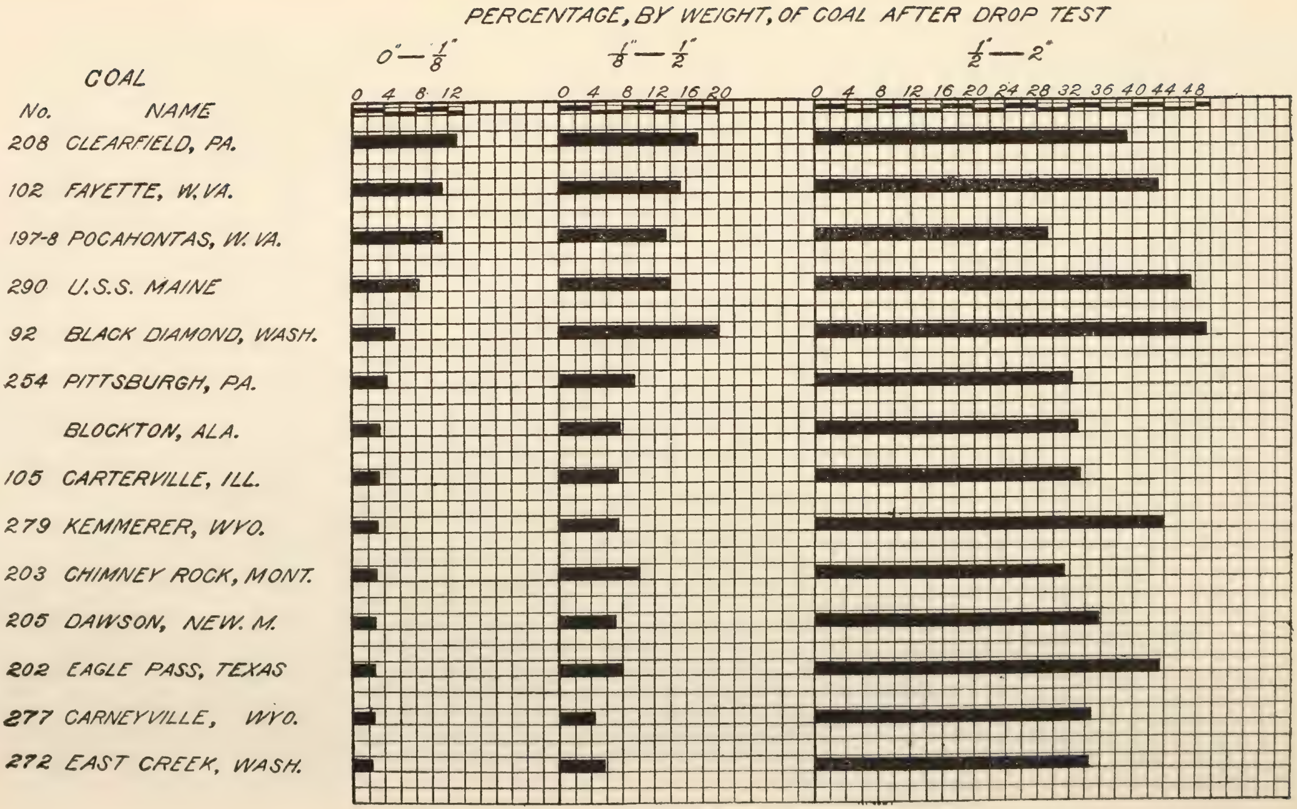
⊛ = IGNITION

A sample of 25 grams of a high sulphur Illinois coal (No. 13451, Vermilion Co., Ill.), 4.38% S, powdered to 60 mesh, was moistened with 25% water and kept under 25 atmospheres oxygen pressure at room temperature for six days. The water soluble sulphur was increased by 0.10% (.025 gram), .05 gram of carbon dioxide was produced, and no sulphur dioxide.

The fact cannot be questioned that iron pyrites, as it occurs in coal, oxidizes in moist air forming FeS_4 and H_2SO_4 and developing heat. This action is known to proceed in practically all coal exposed to the weather. Pure pyrites, also, may develop heat faster than does coal, weight for weight. But, when compared on the basis of the coal as mined, the oxidation of the pyrites present is less than the oxidation of the coal substance itself, and the conclusion appears to be justified that oxidation of the carbonaceous material is the chief factor in starting the initial heating which results in spontaneous fires.

Pyrites is doubtless in many cases a contributory cause. When it occurs in moist, finely divided condition, there may be a pocket of highly pyritic material which will act as an "igniter" to the coal, by raising the temperature just enough to give a stimulus to the coal oxidation. Pyrites also has a mechanical effect in causing, by its weathering, a cleavage of lumps of coal whereby fresh surfaces are exposed. It must be remembered that pyrites requires moisture for its oxidation—theoretically, 15% of its own weight—and that a large proportion of the heat of oxidation is consumed in warming and evaporating the excess moisture present.

The "igniter" action of pyrites pockets appears to be a real element of danger from sulphur in stored coal, but a minor one as compared to the oxidation of the coal substance. The lumps of massive pyrites—the so-called sulphur balls of the miner—are not dangerous, since they oxidized very slowly and only on the surface, the heat thus developed being diffused through the comparatively large mass of the lumps. Porous pockets of wet, finely-divided pyrites are more likely to cause heating. But, in any case, as stated above, the amount of preliminary heating caused by pyrites is greatly lessened by the cooling effect of the moisture necessarily present. The continual accumulation of heat from the oxidation of the finely divided coal itself is the principal element


PERCENTAGE, BY WEIGHT, OF COAL AFTER DROP TEST
0" — 1/8"
1/8" — 1/2"
1/2" — 2"
0 4 8 12
0 4 8 12 16 20
0 4 8 12 16 20 24 28 32 36 40 44 48
COAL
No. NAME
208 CLEARFIELD, PA.
102 FAYETTE, W. VA.
197-8 POCAHONTAS, W. VA.
290 U.S.S. MAINE
92 BLACK DIAMOND, WASH.
254 PITTSBURGH, PA.
BLOCKTON, ALA.
105 CARTERVILLE, ILL.
279 KEMMERER, WYO.
203 CHIMNEY ROCK, MONT.
205 DAWSON, NEW. M.
202 EAGLE PASS, TEXAS
277 CARNEYVILLE, WYO.
272 EAST CREEK, WASH.

of danger. Temperature measurements in coal piles,—examples of which are given later in this paper—show that the initial moderate heating is in most cases evenly distributed through a large area and does not occur in pockets. The subsequent development of high temperature and ignition does, however, take place frequently in pockets owing to irregular conditions of air supply.

The increase of temperature in stored coal beyond the preliminary stage depends on certain physical conditions.

In the first place, temperature is a large factor. The rate of oxidation increases with rise of temperature and the increase is much greater than a linear function. The curve, in fact, as shown in Figure 4*a* approximates the form of a parabola. This curve is based on the data of experiments in which a measured volume of dry air was passed at a fixed rate over powdered, sized, dried coal in an electrically heated tube whose temperature was carefully controlled. The absorption of oxygen was determined by calculation from the initial and final analyses of the air used in the experiment. Five grams of coal (No. 48, Franklin Co., Ill.), 80–100 mesh size, were used in each test, the duration of the test being one hour. Carbon dioxide was produced in these tests in amount not exceeding 1/10 of the oxygen absorbed. It may readily be seen that if coal is given a slight preliminary heating from an external source, its oxidation will be greatly accelerated. Storing coal in warm places, next to furnace walls, over boilers, near steam pipes or waste heat flues, greatly increases the liability to spontaneous combustion.

The second large factor to be considered is dust or fine coal. Since the action of a gaseous substance on a solid is proportional to the extent of their surfaces in contact and since the exposed surface in a given mass is proportional to the fineness of division, it follows that the finer the coal the greater will be the amount of oxidation and the rate of heating in a given mass. Coals differ widely in friability. Figure 5 shows the results of some comparative breakage tests made on lump coals to bring out this point. A 50-lb. sample of screened lump (over 2 inches) was dropped four times from a height of six feet and the coal then sized by screening into three portions, 0″–⅛″, ⅛″–½″ and ½″–2″ The great friability of the low-volatile Appalachian coals is clearly shown and the wide variation between different coal types. It is found in practice that dust and fine coal, when mixed with some larger coal, adds greatly to the danger of spontaneous heating. A single instance from commercial practice will serve to illustrate this. The Calumet & Hecla Mining Co. has three large coal storage piles of 125,000 to 200,000 tons each, in which they store lump coal passed over a $1\frac{3}{4}''$ bar screen just before placing on the piles. No spontaneous fires have occurred since they began this practice, although several fires had been experienced in the same kind of coal before that time.

In the third place, the rate of oxidation of coal is a function of the partial pressure of oxygen present and, therefore, of the rate of flow of air. Experimental results indicating this are shown in Figure 4*b*. The amount of air admitted to the interior of a coal pile influences in large measure the heat development. If the admission of air to the pile is restricted by piling only uniformly fine coal or by banking up the sides with tight bulkheads, the accumulation of heat soon becomes so slow as to be balanced by the losses from radiation and convection. The percentage oxygen in the air becomes reduced to a point where its action on the coal is very slow.

Some observations made on a large commercial stock pile illustrate these points (Figure 6). The chart shows temperatures at different depths in the pile and the composition of air samples withdrawn from corresponding depths. Where the coal is warm the oxygen in the air is below normal and in some such places (as at E, Figure 6) the oxygen is very low, although the coal is only slightly heated. The coal at point E was fine and contained very few lumps, so that the penetration of air was restricted. At D, Figure 6, on the other hand, there was evidently a strong draft of air from below causing a pocket of fire at 20-23 feet depth. The observations at points A and C illustrate the uniformity of heating throughout a large mass in the early stages of heat development.

Cases are known in commercial practice where it was found that a large pile of uniformly fine slack coal gave only slight preliminary heating and no serious fires, while the same coal when stocked as run-of-mine with a large percentage of fine, gave very serious trouble with fires. When the amount of air supply will support oxidation at a rate sufficient to counterbalance the heat carried away by the air and by other losses, there will result a gradual and continuous rise in temperature.

The fourth factor to be mentioned as influencing the development of heat is the degree of freshness of the coal surfaces. Freshly mined coal is known to absorb oxygen with greater avidity than coal which has been exposed for some time. Figure 4*c* gives experimental results showing the oxygen absorbed by the same coal sample in three successive treatments with a current of air at 140° C. The marked reduction in oxygen absorbed after the first treatment is evident. In practice it is found that freshly mined or freshly broken coal, when stored without a preliminary period of "seasoning," gives serious trouble from heating. Also, if coal is rehandled after six weeks or two months and thus thoroughly cooled, it rarely heats again spontaneously.

Some coals by their chemical nature are more liable than others to spontaneous heating, but any bituminous coal will heat if the storing conditions are such as favor heating. Large piles (roughly more than 12 feet in depth) involve danger, since the possibility of dissipation of the accumulating heat becomes less as the bulk increases. Either screened lump coal or a uniformly fine slack is safer for storing than run-of-mine. Friability in coal is a very undesirable quality. Pyrites may contribute largely in some cases to spontaneous heating if it occurs in moist, finely divided condition, more or less accumulated in pockets. It is probably, however, in most cases a minor factor.

A NEW METHOD OF IGNITION FOR BOMB CALORIMETERS WITH SPECIAL REFERENCE TO COAL CALORIMETRY

By Victor H. M. Roehrich
Municipal Testing Laboratories, St. Paul, Minnesota

For careful work the bomb calorimeter is usually given the preference in coal testing but it is desirable to have it arranged as conveniently as possible for technical use. An improvement that helps to simplify the work is the use of a platinum fuse wire. This does away with the correction for heat of combustion necessary with iron wire. Furthermore, as arranged in my design the same wire is used repeatedly and remains attached to the combustion pan. Its free end need only be slipped into the hook provided for that purpose, whereupon it is ready for use. The Emerson Calorimeter uses a fine platinum fuse wire but this melts in the first determination. Furthermore, as the makers state, it cannot be used in standardizing, as with benzoic acid, etc.[1] The purpose of the present paper is to show how these objections have been obviated.

Tests were made to determine whether a coarser wire could be used so as to make it possible to employ the same wire repeatedly, but it was found that even a No. 16 platinum wire, used as a supporting terminal, melted if placed over the burning coal. The next step was to place the fuse (F) off to one side in a channel (C) communicating with the body of the platinum pan (P). See figure. The fuse is a two cm. piece of No. 28 platinum wire; however, only about one cm. of it carries current. One end is fastened through two holes in the pan (h) and (h^1). The other end is bent to slip into the platinum hook (H) of No. 16 wire which forms one of the electric terminals. The current passes from (H) through the fuse and pan and to a ring-shaped pan support forming the other terminal. The current is taken from a six-volt storage battery and a resistance of 75 inches of No.

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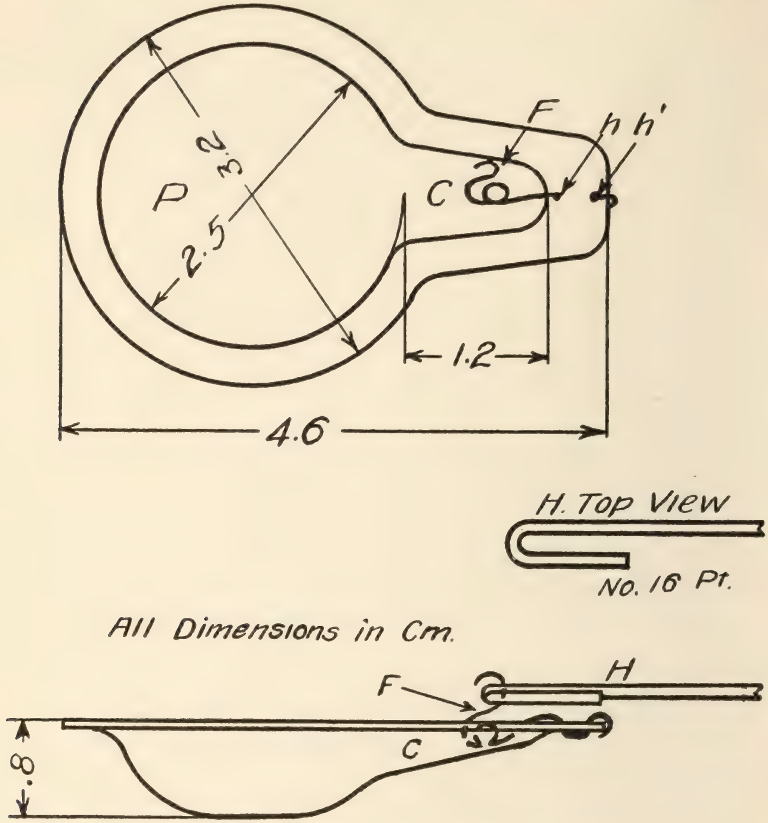
[1] Pamphlet Emerson Apparatus Co., Boston, 1909.



18 iron wire is interposed in series with the fuse. Contact is made by a spring key. The current is left on just long enough to heat the fuse red hot, which can be determined by trial. We have found two seconds to be the proper time. Each contact introduces sufficient heat to raise the temperature from .0027 to .0034° C. (obtained by making ten contacts in succession). This means a total of about seven calories with a probable variation or error of two calories in each determination, which is close enough for almost any calorimetric work.

Bituminous coals are easily ignited, a little of the coal being placed in the channel to cover the coil of the fuse wire and carry the flame to the body of the pan. Anthracite coals can also be successfully run, but in this case the pan is lined with a paste of asbestos moistened with ether and alcohol, which is dried and ignited and weighed. This is a satisfactory method of applying the asbestos to insulate the anthracite and insure complete combustion.[1]

As previously stated, a platinum fuse wire is not suitable for standardizing. This is because the naphthalene or benzoic acid melt and volatilize and run off the fuse before it is red hot, at the same time retarding the heating of the wire by their heat absorption. Standardization may be accomplished by igniting the standard material with iron wire and then adding to the observed rise in temperature .003° C. or the rise due to the platinum fuse so as to allow for this rise in the regular determination where the platinum fuse is used. Of course the rise due to one contact through the platinum fuse must be determined for each individual calorimeter.

It has been found , however, that by covering the platinum fuse wire with asbestos containing a little powdered benzoic acid or naphthalene, ignition readily occurs, first because the fuse is not cooled as much as if in the pure melted substance, and second because the material is prevented from running off of the fuse before ignition takes place The procedure is to fill the channel in the fuel pan with about 10 mg. asbestos, so as to cover the coil of the fuse wire. Ignite, weigh the pan and asbestos and pour in the naphthalene or benzoic acid which has been barely melted in a small platinum crucible. Avoid allowing the melt to run into the asbestos. The nearly solidified material is drawn up to almost touch the asbestos by means of a platinum wire. Now a small amount of pulverized napthalene or benzoic acid is mixed with the asbestos in the channel by means of a pointed spatula, being careful not to remove any asbestos from the pan. Cool in a desiccator and weigh, getting the weight of substance to be burnt. By melting the material and pouring into the pan, the use of a pellet press is avoided, but if desired a pellet of the material to be burnt may be laid next to the mixture of asbestos with pulverized substance.

[1] A. G. Blakely and E. M. Chance have recommended the use of asbestos, *J. Ind. Eng. Chem.* *3*, 557, 1911.

The corrected water equivalent of our calorimeter run with iron wire and with correction for use of platinum fuse was 2357.3 Cal. based on a number of tests. Two tests were now run using the platinum and asbestos. The one with standard napthalene gave 2355 and the second with standard benzoic acid gave 2356.

Oils can also be ignited by weighing into the pan and then adding a suitable amount of ignited asbestos to cover the fuse, mixing a little oil with it by means of a short platinum wire which is left lying at the side of the channel during combustion to burn off the oil adhering.

There seems a possibility that the proposed design of pan and fuse can also be used in general in more scientific calorimetric work using asbestos around the fuse for liquid or easily melted substances. It has the advantage of greater simplicity and does away with the deterioration of the platinum that results if iron wire is used.

ON THE FORMATION OF ALDEHYDES BY OXIDATION OF MINERAL OILS

By Ferd. Schulz
Bohemian Polytechnical High School, Prague, Bohemia

The oxidation of mineral oils has been the subject of repeated studies. The chief investigations, due to Pouchet, Gill a. Memel, Willigk, Hausmann a. Pilat, Charitschkoff concerned the formation of acids and aromatic hydrocarbons. In this preliminary notice I will call attention to a new method, by means of which both the liquid mineral oils and the solid paraffin can be easily oxidized, aldehydes being the chief product of the reaction.

The mode of procedure is as follows. A tubulated glass retort is filled with melted paraffin and the tubulus is connected with a series of Drechsel's washing bottles by means of a T tube, through which a thermometer is inserted, dipping into the liquid. After the paraffin has been heated to 290-300 degrees centigrade (thermometer in liquid) a rapid current of air is drawn through the apparatus by a water air pump. The reaction immediately begins with a rather violent explosion, the vapours of paraffin take fire and burn during some seconds within the retort, whereupon a thick white smoke passes through the washing bottles.

The operation can be then carried on during many hours without any danger of explosion, if only the liquid is kept always simmering gently at 290-300 d. centigrade and care is taken for a sufficient strong current of air. Through the neck of the retort a small quantity of paraffin is distilling, while the largest part of the vapours is drawn by the current of air and yields a white smoke, possessing a pungent, irritating smell.

The paraffin becomes yellow, then brown and black; after the whole content of the retort has been oxidized, there remains about 3% asphalt of a very high melting point.

When filling the washing bottles with water, we obtain the following products:

1. A semi-solid yellow mass of crystals, which after squeezing

out the adherent liquid and washing with a mixture of alcohol and ether, yields white, odourless scales, probably the unaltered paraffin. The oil, which has been drawn off, has a suffocating smell and shows the spec. gravity 0,868 at 15 d. C. and the viscosity 1.2° Engler; it begins to distill at 152 d. C. and 52% passed over between 152 and 300 d. C.

A large part of the oil dissolves in a solution of hydrogen sulphite of sodium, and it shows the general characteristic of aldehydes on examination with Schiff's reagent, Fehling's solution and alkaline silver solution. On addition of phenylhydrazine the oil becomes warm, and a red precipitate is thrown down, not soluble in ether. A dilute solution of caustic soda takes up a small quantity of acids, which have the unpleasant smell of petrolacids and show the Charitschkoff's reaction with copper sulphate and benzine.

2. The wash-waters, possessing a very irritating smell, which resembles one at the same time of acrolein and formic aldehyde, showed a very strong reaction with alkaline silver solution and the reagents of Schiff, Schryver and Ganassini. On saturation with sodium chloride the water was distilled, till 10% of it passed over, and the distillate obtained, saturated with sodium sulphate after calcium carbonate was added to neutralise the acids, was submitted to a new distillation from a water-bath.

A colourless, mobile liquid passed over between 60 and 83° C. and was separated by fractional distillation into chief fractions, boiling between 39-46° C.; 46-52° C.; 52-65° C.; 65-83° C.

In identification of individual substances I encountered considerable difficulties, very few definite compounds of the fatty aldehydes being on record. This phase of the investigation is to be prosecuted further.

3. The gases, passing through the washing bottles, were examined in a gas burette, and the following composition was found at different phases of the procedure:

Carbon dioxide	0,2-0,4%
Heavy hydrocarbons	0,0-2,1%
Oxygen	9,0-17,1%
Carbon monoxide	0,3-5,0%
Light hydrocarbons (calculated as methane)	1,2-4,2%

My further investigations proved that the lubricating oils, produced from Boryslav-oil, can be oxidized in the same manner.

Conclusion: The vapours of solid paraffin or lubricating oil, produced from the Boryslav-oil, when oxidized by a current of air, yield chiefly aldehydes, carbon monoxide and a small quantity of acids.

DE L'ALTERATION DES POUSSIERES DE HOUILLE EXPOSEES A L'AIR

PAR M. TAFFANEL

Directeur de la Station D'Essais de Liévin. Pas de Calais, France

L'altération de la houille exposée à l'air a déjà fait l'objet de nombreuses études; on connaît notamment les remarquables travaux de H. Fayol, qui datent d'une trentaine d'années, mais sont toujours consultés avec fruit par ceux qui s'intéressent à cette question.

Mais Fayol et les auteurs qui l'ont suivi ont eu surtout en vue le problème de l'altération des houilles en stock, de leur dépréciation comme combustible, de la diminution de leur aptitude à donner du coke de bonne qualité; ou bien ils ont envisagé cette dangereuse propriété qu'ont certaines houilles de s'échauffer en plein massif et de donner naissance à des feux souterrains.

La station d'essais de Liévin a repris la question à un point de vue différent. Du moment que les houilles sont capables, dans certaines conditions, de s'altérer sous les influences atmosphériques, n'en peut-il être de même des poussières charbonneuses qui tapissent les galeries de mine? Il est vrai que la poussière, dispersée sur le sol et les parois, ne saurait subir cet échauffement qui favorise l'altération au sein d'un tas ou d'un massif; mais d'autre part la grande finesse des grains de poussière a pour effet de considérablement augmenter les surfaces sur lesquelles l'air exerce son action.

Si les poussières des mines subissent réellement une altération sensible, une importante question se pose. A la Station d'essais de Liévin et dans les sièges d'expériences des autres pays, Pittsburg. Bruceton, Altofts, Frameries, Gelsenkirchen, Rossitz, on étudie lesrisques d'explosion des poussières de houille et les moyens d'empêcher le retour des trop fréquentes catastrophes qui dévastent les exploitations minières; les grandes quantités de poussières nécessaires pour les expériences sont obtenues par broyage de charbon relativement frais. Ces poussières artificielles ne subissent l'action de l'air que pendant un petit nombre de jours, tandis que les poussières de la mine sont exposées parfois pendant des mois, ou même des années, aux influences atmosphériques. L'altération agit-elle sur les propriétés dont dépend le degré d'inflammabilité? Aggrave-t-elle ou atténue-t-elle le danger d'inflammation? Les conclusions tirées de l'étude des poussières artificielles tendent-elles à exagérer le danger, et dans quelle mesure, ou au contraire, ce qui serait plus grave, risquent-elles de doner l'illusion d'une sécurité trompeuse?

De la solution de ces questions dépend le degré de confiance que l'on doit accorder aux mesures préventives prescrites, à la suite des essais en galerie, pour la prévention des catastrophes minières. Le problème présénte donc un intérêt capital au point de vue de la sécurité dans les mines. En même temps les recherches entreprises pour le résoudre apportent des renseignement nouveaux et intéressants au point de vue de l'étude des combustibles.

L'altération des poussières de houille a été étudiée à la Station d'essais de Liévin par diverses méthodes dont il convient de dire quelques mots avant de passer à l'exposé des résultats.

On y observé, comme Fayol en 1875, les variations de poids par exposition à l'air à la température ordinaire, et aussi, pour quelques essais de comparaison, à des températures plus élevées.

Puis, pour distinguer les diverses causes d'augmentation et diminution de poids, qui, en se combinant, donnent comme résultante la loi de variation de poids observée, on a étudié les echanges gazeux en enfermant de petites quantités de poussières dans des ballons remplis d'air, scellés, puis ouverts, pour analyse du contenu gazeux, après des temps variables. Il faut avoir soin de prendre des ballons assez grands pour que les échanges gazeux qui se produisent au cours de l'essai ne modifient pas trop la composition de l'atmosphère; d'ailleurs on peut étudier l'influence du confinement en faisant systématiquement varier le rapport du poids de la poussière au volume du ballon.

Ensuite on a exposé de la poussière fraîche à l'atmosphère de la mine, dans des boîtes spéciales combinées de manière à assurer autant que possible l'intégrité de l'échantillon étudié, sans intrusion de particules étrangères; on a suivi, par des prises d'essai, les variations de composition de la poussière mise en observation.

Enfin on a complété ces études systématiques par l'analyse de divers échantillons prélevés dans les galeries de mine. On ne s'est pas contenté d'une analyse globale, mais on a divisé chaque échantillon en ses éléments suivant la finesse et la densité et chaque élément a fait l'objet d'une étude spéciale.

Voici maintenant les résultats de ces travaux.

La première variation de composition qui se manifeste, quand la poussière de houille a pris naissance au chantier d'abatage, est un dégagement de formène. On sait que ce dégagement se continue assez longtemps, lorsqu'il s'agit de fragments aux dimensions commerciales. Il importait de connaître l'importance et la durée du dégagement pour les fragments pulvérulents. La multiplication des surfaces accélère sans aucun doute le dégagement; mais celui-ci est loin d'être immédiatement complet. Des échantillons de charbon fraîchement abattu dans une mine grisouteuse ont dégagé, pendant les premières heures qui ont suivi la pulvérisation, de 0,4 à 1,4 centimètre cube de formène par gramme de charbon et par heure; la vitesse de dégagement n'a pas tardé à se ralentir, vers la fin du premier ou du quatrième jour; suivant les cas, son ordre de grandeur fut le vingtième de centimètre cube par heure et par gramme de poussière; vers le 5ème ou 10 ème jour la première phase de dégagement relativement rapide était terminée; un régime lent commença, qui se poursuivit pendant un grand nombre de jours; l'ordre de grandeur de la vitesse de dégagement était alors de quelques millimètres cubes par jour et par gramme, ou moins encore au bout d'un certain temps; sur de vieilles poussières ayant séjourné quelques mois au fond de la mine, le dégagement de formène n'a pu être décelé. C'est, en définitive, pendant les dix premiers jours que se dégage la plus grande partie du formène libérable; sur les échantillons étudiés, la quantité libérée pendant cette période a atteint 14 à 22 centimètres cubes par gramme; ces chiffres ne comprennent pas le formène dégagé du morceau de houille pendant les quelques heures qui ont séparé l'abatage de la pulvérisation, ni celui que la pulvérisation, rapidement menée, a libéré. Les résultats que nous venons de résumer se rapportent à de la poussière très fine, ne donnant que peu de refus au tamis n°200; du menu plus grossier, provenant de la même exploitation, donnerait un dégagement plus prolongé et, au total plus important, car il aurait perdu moins de grisou à la pulvérisation; mais, au point de vue de l'inflammabilité des poussières, ce sont les parties les plus fines qui offrent le plus d'intérêt. On remarquera que si un nuage tenant 500 grammes de poussières au mètre cube d'air, taux convenable pour l'inflammation, est constitué par des poussières fraîches, il peut fournir à la combustion un appoint de gaz combustibles supérieur de 1 litre par mètre cube environ à ce que pourrait donner une poussière ayant passé la première phase du dégagement. Une inflammabilité au moins égale serait obtenue avec un nuage de cette dernière poussière en suspension dans de l'air tenant 0,1% de grisou. Or les essais montrent qu'une telle proportion est sans effet appréciable sur l'inflammabilité. Donc si, en principe, la petite quantité de grisou spontanément libérable, que les poussières fraîches n'ont pas encore dégagée, est plutôt de nature à les rendre plus inflammables que les poussières moins fraîches, qui servent dans les galeriés d'expériences, cet effet doit être considéré, dans la pratique, comme absolument négligeable.

L'effet le plus important de l'exposition des poussières à l'air est l'oxydation; cet effet se manifeste sous deux formes: fixation d'oxygène sur la houille, départ du carbone à l'état d'acid carbonique; il se peut qu'il y ait un troisième effet consistant en départ d'hydrogène à l'état de vapeur d'eau; mais on n'a pu le déceler au degré de précision des essais; de même les quelques déterminations d'azote, effectuées avant et après altération, n'ont révélé aucune modification sensible de la proportion de ce corps.

De ces divers effets, c'est la fixation d'oxygène qui constitue le phénomène prédominant. La vitesse de fixation varie avec la nature du charbon; elle paraît en général plus grande pour les charbons riches en matières volatiles que pour les charbons pauvres en ces matières; elle varie avec le temps, assez rapide au début de l'exposition, plus lente ensuite.

Tout à fait au début de l'exposition, un charbon pulvérulent à 30% de matières volatiles fixe 0,2 centimètre cube environ d'oxygène par gramme de charbon et par jour; le volume total fixé atteint 1 centimètre cube au bout de 10 jours, 2 cc. au bout de 30 jours, 3 cc. au bout de 60 jours, 4 cc. au bout de 120 jours. On remarque que le poids fixé d'oxygène est inférieur pendant les dix premiers jours environ, et supérieur ensuite au poids de formène dégagé; c'est pourquoi le poids de l'échantillon diminue d'abord puis augmente; sur un échantillon moins frais, on ne constaterait que l'augmentation de poids.

Avec des poussières moins riches en matières volatiles, la vitesse de fixation est deux ou trois fois plus faible.

Enfin des poussières prélevées au fond de la mine et datant de deux à quatre mois continuent à s'oxyder, en fixant 1 centimètre cube d'oxygène par gramme, en 110 à 140 jours.

Les résultats ci-dessus ont été déduits des essais faits en ballons scellés, en présence d'air assez sec et pur au début de l'essai, mais s'appauvrissant jusqu'à ne tenir, à la fin de l'essai, qu'environ 16% d'oxygène. Dans la mine l'air, quoiqu'un peu vicié, conserve des teneurs en oxygène plus élevées (19 à 20% aux deux points où de la poussière fut mise en observation); l'humidité est parfois assez prononcée. Pour ces raisons et d'autres encore peut-être, l'oxydation est parfois plus active; on a constaté qu'après 100 jours d'exposition un gramme de poussière de houille à 30% de matières volatiles fixait 7 centimètres cubes d'oxygène dans un cas, 27 centimètres cubes dans un aètre cas; le second cas correspond à un retour d'air particulièrement humide. Ces chiffres sont deux à sept fois plus élevés que ceux obtenus en ballons scellés; ils correspondent à des augmentations de teneur en oxygène respectivement égales à 1,0 et 3,9%.

Le départ de carbone à l'état d'acide carbonique peut être décelé dans les ballons scellés; mais la quantité dégagée demeure très faible; il a généralement fallu de 75 à 150 jours pour obtenir le dégagement d'un dixième de centimètre cube par gramme; la perte de poids ou de teneur en carbone correspondant à ce dégagement est absolument insignifiante; un seul échantillon, prélevé au fond de la mine et déjà fortement oxydé, a dégagé 0,5 centimètre cube d'acide carbonique en 150 jours; c'est à peu près la moitié du volume d'oxygène fixé pendant le même temps; le dégagement d'acide carbonique n'est donc plus négligeable, mais la fixation d'oxygène n'en reste pas moins le phénomène prédominant. Ainsi la combustion lente du charbon exposé à l'air, qui devient très importante lorsqu'on échauffe la poussière, s'est montrée généralement insignifiante et en tous cas peu importante à la température ordinaire; on n'a pas rencontré de cas où elle ait été suffisante pour déterminer une diminution de poids de la poussière.

Les poussières recueillies dans les galeries de mine sont des mélanges complexes où l'on trouve des fragments de houille, de roches du gisement, de schistes charbonneux, et aussi des débris de matières organiques diverses; on a séparé ces derhières, qui sont riches en oxygène et matières volatiles; leur proportion est généralement assez faible pour qu'elles ne modifient que fort peu les propriétés de l'ensemble. L'oxydation est généralement fort prononcée; la teneur en oxygène, rapprochée de ce qu'elle devrait être pour des mélanges de roche et houille fraîche, accuse des différences en plus allant jusqu'à 8% pour la poussière brute et jusqu'à 15% lorsqu'on les rapporte à la partie houille, la seule qui subisse l'oxydation. Quelques cas particuliers, étudiés avec plus de détail, ont montré que les grains de houille pure, dont on connaissait l'origine et qui dataient de deux à quatre mois seulement, étaient peu oxydés; leur degré d'oxydation correspondait à ce que l'on pouvait prévoir d'après les essais ci-dessus relatés; mais les schistes charbonneux étaient profondément altérés; l'explication est la suivante: les fragments pulvérulents de schistes charbonneux, qui entrent dans la composition des poussières proviennent de la dégradation progressive des parois des galeries sous l'influence des agents atmosphériques ou du piétinement; les bancs de schistes charbonneux sont en effet laissés en place et non exploités; plus tendres que les autres roches stériles laissées en place, ils contribuent pour une part relativement grande à la formation des poussières; les fragments de schistes peuvent n'exister à l'état pulvérulent que depuis peu de temps; comme dans les exemples étudiés, où les galeries avaient été soigneusement dépoussiérées deux à quatre mois auparavant; ils n'en avaient pas moins commencé à subir l'altération de l'air, sur les parois de ces galeries, depuis l'époque du creusement, c'est-à-dire depuis plusieurs années; c'est pourquoi ils manifestent une oxydation aussi importante.

Le degré d'oxydation des poussières de la mine ne dépend donc pas seulement de la nature de la houille exploitée, de sa friabilité, de la température ou de l'humidité de l'air et des multiples circonstances qui font que les particules charbonneuses déposées en tel ou tel point sont de pulvérisation plus ou moins ancienne il dépend aussi de la nature des parois des galeries, de l'époque de leur percement et des réparations qu'elles ont subies, de la proportion de schistes charbonneux qui s'y rencontrent; en sorte que, si l'on peut affirmer qu'habituellement les poussières de la mine sont plus ou moins oxydées, on ne peut fixer de règle simple qui permette de prévoir, dans chaque cas, le degré d'oxydation.

Deux remarques achèveront de caractériser l'altération des poussières.

La teneur en matières volatiles de la houille oxydés est plus élevée que celle de la houille fraîche, en sorte que dans les mélanges recueillis au fond, cet accroissement de teneur fait souvent plus que compenser la diminution de teneur due à la partie rocheuse, pauvre en matières volatiles. Le pouvoir calorifique d'un poids donné de poussières est plus faible quand les poussières sont altérées; car dans ce dernier cas, l'appoint d'oxygène a pour effet de diminuer la proportion du carbone et de l'hydrogène, qui sont les éléments actifs de la combustion. La diminution de pouvoir calorifique semble même assez grande pour qu'il faille admettre que la fixation de l'oxygène sur le charbon est une réaction exothermique, comme on pouvait vraisemblablement le supposer; mais le degré de précision des déterminations calorifiques n'a pas permis de préciser la chaleur dégagée dans cette réaction.

La preuve étant faite que les poussières déposées dans les mines étaient oxydées, il devenait important de comparer les propriétés des poussières fraîches et altérées au point de vue de l'inflammabilité.

Divers essais ont conduit à penser que, dans le mécanisme de la propagation de la combustion dans les nuages de poussières, un rôle important appartenait aux matières volatiles dégagées pendant la phase d'échauffement qui précède la phase de combustion vive. En conséquence on a recherché s'il existait des différences dans la nature et la quantité des produits volatils dégagés par les poussières fraîches ou altérées, spécialement pendant les premières phases de la distillation. Une nacelle contenant 5 grammes de la poussière à étudier est placée dans un tube de porcelaine de Saxe, vernissée intérieurement. Le tube est chauffé par un fort chalumeau à gaz; un couple thermoélectrique, plongé au sein de la poussière, permet de suivre l'élévation de température; on fait le vide avant le début de l'échauffement; puis les gaz sont recueillis sur la cuve à eau par un tube de dégagement; on recueille et analyse séparément les produits dégagés pendant diverses phases successives de l'échauffement. L'allure du chauffage est telle que la température au sein du charbon atteint 500 degrés centigrades en 2 minutes, 1000 degrés en 10 minutes, et 1200 degrés en 15 minutes; le dégagement gazeux est assez rapide au début, puis se ralentit progressivement. On a comparé de la poussiére fraîche et des poussières plus ou moins oxydées, provenant de la même mine. Les résultats sont résumés dans le tableau ci-joint.

La distillation commencevers 450 à 500 degrés pour la poussière fraîche, vers 550 à 600 degrés pour les poussières oxydées; les quantités dégagées pendant les premières étapes de la distillation diffèrent beaucoup; 100 grammes de poussières donnent, jusqu'à 800 degrés, 22 litres de gaz pour l'échantillon frais, et 10 litres environ pour les échantillons altérés; il est vrai que ces derniers étaient plus cendreux; en rapportant à 100 grammes, cendres déduites, on obtient 24 litres d'une part, 14 à 15 litres d'autre part; le volume dégagé pendant cette première phase atteint, pour les poussières fraîches, les deux tiers du volume total que dégage la distillation complète; il n'atteint même pas le tiers pour les poussières oxydées.

La composition des gaz recueillis dans les deux cas est également très différente, comme le montre le tableau suivant:

		Carbures Éthyléniguet	Méthane	Hydrogène	Oxyde de Carbone	Acide Carbonique	Oxygène	Azote
		%	%	%	%	%	%	%
Gaz dégagés avant 800°	Poussière fraîche	2,9	29,8	51,8	4,9	1,25	1,25	8,10
	Moyenne de poussières oxydées...	1,7	26,7	41,9	12,2	9,0	1,5	6,8
Produits gazeux de la distillation complète	Poussière fraîche	1,9	22,6	56,4	9,2	1,3	1,1	7,5
	Moyenne de poussières oxydées ...	0,6	12,8	51,0	24,0	3,9	0,9	6,7

Les produits à basse limite d'inflammabilité, qui jouent le rôle le plus actif dans la propagation de la combustion, à savoir les carbures éthyléniques, le méthane et l'hydrogène, sont en proportion plus faible pour les poussières oxydées que pour les poussières fraîches; au contraire l'oxyde de carbone, à haute limite d'inflam mabilité, est plus abondant pour les poussières oxydées; il en est de même de l'acide carbonique, qui tend à s'opposer à la combustion.

Plusieurs causes s'unissent donc pour rendre les poussières altérées moins inflammables que les poussières fraîches; leur pouvoir calorifique est moindre; la distillation des matières volatiles commence à une phase plus tardive de l'échauffement et est, au début, beaucoup moins active; les gaz distillés sont, pour diverses raisons, moins inflammables.

On a vérifié que les poussières oxydées étaient en effet moins inflammables que les poussières fraîches.

L'essai a été fait au moyen d'un appareil de laboratoire combiné pour l'étude des poussières au point de vue de l'inflammabilité. Un décigramme de poussières placé dans un tube de soufflage est projeté en nuage à travers un tube de procelaine de 25 millimètres de diamètre intérieur et 10 centimètres de longueur, placé l'axe vertical, et chauffé à environ 1000 degrés par un chalumeau ou un four électrique; à partir d'un certain degré inflammabilité, le nuage prend feu et donne à la sortie du tube une flamme plus ou moins volumineuse, que l'on photographie; les dimensions de la flamme caractérisent le degré d'inflammabilité de la poussière et l'on a vérifié qu'il y avait, d'une manière générale, accord entre les classements obtenus par cette voie et ceux que donnent les essais d'explosion dans la galerie d'expériences.

Comme les poussières oxydées ne peuvent être recueillies avec de suffisantes garanties d'homogénéité, en assez grande quantité pour être expérimentées dans la galerie, elles ont été comparées aux poussières fraîches dans l'appareil de laboratoire. On a observé une différence systématique d'inflammabilité; si t représente la teneur en cendres d'un échantillon de poussières oxydées prélevées au fond de la mine, il aurait fallu, pour obtenir une flamme de même grandeur avec des poussières fraîches, additionner celles-ci de roches pulvérisées jusqu'à ce que l'on atteigne une teneur en cendres supérieure à t, et atteignant parfois $t+10$ ou $t+20$; ainsi l'oxydation avait diminué l'inflammabilité comme l'aurait fait l'addition de lo ou 20 unités pour cent de poussières incombustibles.

On conclut de ces recherches que lorsque, dans les galeries d'essais où l'on étudie les explosions de poussières, on emploie pour les expériences des poussières artificielles, on se place dans des conditions telles que les explosions sont plus faciles ou plus violentes que si, toutes choses égales d'ailleurs, on employait des poussières naturelles. Il n'y a donc pas à craindre que, du fait du choix des poussières, les conclusions que l'on en tire pour la prévention des accidents risquent d'être mises en défaut dans les conditions du fond de la mine. Par cette constatation, se troube rempli le but principal que poursuivait la Station d'essais de Liévin en entreprenant ces recherches.

Les résultats obtenus apportent en outre une contribution à l'étude générale de l'oxydation des houilles.

COAL TAR LIGHT OIL IN THE UNITED STATES: THE MANUFACTURE, NATURE AND USES OF PRODUCTS DERIVED THEREFROM

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Introductory

This paper is an attempt to give a rather general outline of that part of coal tar refining dealing with the lighter oils obtained in the first distillation of coal tar. The writer has taken as his theme the so-called "coal tar light oil," and has, in the general description, written as if this were the only source of the refined products mentioned. It must not be forgotten that other products are used as raw materials for the same purposes, and are mixed with light oil products at various stages of manufacture. Thus coal tar carbolic oil, water gas tar light oil, crude benzol from washing coke oven gases, and Pintsch gas drips may serve as raw materials to be introduced at some point or other in the manufacturing processes.

The figures given for production include products from all these sources, and are not confined to light oil products alone. The figures are the result of compiling as much information as the writer was able to obtain from various manufacturers.

Light Oil Production

Light oil is produced in the first distillation of coal tar, being the first fraction of oil taken off. In practice, it usually consists of that portion of oil collected while the water is being removed. The amount of light oil obtained from different tars varies considerably, some containing only a trace, while in others it may run as high as 4%. A fair average for coal tars distilled in the United States is about 2.0%.

Nature of the Material

Light oil of coal tar, as produced in the United States, varies in composition, depending on the nature of the tar from which it is produced, and upon the method of distillation used. The variations in composition are rather more of proportion of constituents than of kind; in other words, in all coal tar light oil we meet with the same bodies, but their amount can vary quite widely.

The main constituents of coal tar light oil can be roughly grouped into four classes:

1. Neutral hydrocarbons of the aromatic series.
2. Unsaturated hydrocarbons of various series, the main one, however, being of the styrolene type.
3. Basis bodies, consisting of pyridine and the higher homologues.
4. Acid bodies known as "tar acids," in light oil consisting of phenol and the three isomeric cresols, with small amounts of the higher homologues.

Besides bodies falling into these classes, there are small amounts of other materials such as carbon bisulphide, various organic nitriles and various sulphur compounds, such as thiophene, etc. All the materials manufactured from light oil in the United States fall in the first and fourth classes mentioned above.

Methods of Testing Light Oil

The assay of light oil usually consists in:

1. A distillation, which gives the approximate amount of hydrocarbons boiling at various temperatures.
2. Determination of tar acids.
3. Determination of tar bases.
4. Determination of naphthalene.
5. Determination of specific gravity.

These tests, with the exception of that for tar bases, have been described in detail, with drawings of the apparatus used, by Mr. S. R. Church (Journal of Industrial and Engineering Chemistry, Volume 3, No. 4, April, 1911). The tar base test is carried out in exactly the same manner as the test for tar acids, preceding this in the analytical process, with the exception that 20° Beaume sulphuric acid replaces the 10% soda used in the corresponding tar acid test.

In exceptional cases more exhaustive tests are made, which include a very careful fractionation of the light oil neutral hydrocarbons, to determine the approximate percentage of pure benzene and toluene, etc., but as these are but seldom used, it seems well to omit them here.

As a means of showing how widely light oil from various sources can vary, the following tests are given as representing average coal tar light oil produced in the United States:

	Maximum	Minimum	Average
Specific gravity	1.002	0.953	0.985
Tar acids	18.0%	4.0%	7.5%
Naphthalene	29.9	trace	13.2
Hydrocarbons boiling below 200° C	62.0	30.0	48.0

These tests do not include the determination of tar bases, as this is seldom made. I may say, however, that the bases amount to but one to three per cent of the light oil.

First Distillation of Light Oil

This is carried on in iron horizontal cylindrical stills, with a capacity of from 3000 to 5000 gallons, in size varying from six to eight feet in diameter, and fourteen to sixteen feet in length. The stills are set in brick work with either oil or coal firing. The type of setting has changed from time to time, the most approved type at present being that with a firebrick arch, the hot gases of combustion passing under the arch to the back and then over it under the still to the stack at the front. The stills are provided at the top with a vapor pipe, an opening for charging and a manhole for cleaning, at the bottom with a draw-off for removal of the residue. The vapor pipe runs directly to the condenser without any baffling or fractioning device intervening. A thermometer set in the vapor pipe of the still serves to regulate the fractions taken off. The distillate is run directly from the condenser into tanks or pans, depending on its nature.

In this first distillation, the light oil is split into but two fractions, one, crude naphtha, up to about 200° centigrade, taken off into a storage tank, and above that point, napthalene oil, which is run directly into large, shallow iron pans. The residue in the still is thick, and is run out when cool and mixed with crude tar.

The heavier fraction, containing naphthalene, is allowed to cool to air temperature, when the naphthalene settles out. As much oil as possible is drained off by gravity, and the drained naphthalene centrifugalled to further remove oil. These separated crystals form crude naphthalene, the working up of which will be described later. The oil drained from the naphthalene is moderate boiling creosote, containing a high percentage of tar acids, almost entirely cresols and the next higher homologues. This oil is used as the basis of "percentage" carbolic acids second quality, as described later, or is artificially frozen to further remove naphthalene, and the frozen, well settled oil, containing about 25% of tar acids, is used for the manufacture of disinfectants and sheep dips, or as a basis for first quality "percentage" carbolic acids.

Treatment of Crude Naphtha

The crude naphtha is first brought into iron cone-bottom agitators of about 12,000 gallons capacity, and the tar acids removed by treatment with dilute caustic soda solution. This solution of tar acids (mainly phenol and cresols) serves as the raw material for the manufacture of the refined grades of carbolic acid, as will be described in a later part of this paper. The naphtha freed from tar acids is then treated in lead-lined agitators with sufficient dilute sulphuric acid to remove basic bodies. At present, owing to the fact that the market for pyridine bases is very limited in the United States, no attempt is made to recover these bases, but the pyridine sulphate liquor is thrown away.

The extracted naphtha is then subjected to fractional distillation under vacuum of about 29 inches, using a column. This is carried out in stills of the same size and type as used in the first distillation, a column and reflux condenser being used between the still and the condenser. The fractions are collected of necessity in closed receivers. This distillation is regulated, like the other, by a thermometer in the vapor pipe. Four fractions are taken off:

1. Crude benzol and toluol fraction.
2. Intermediate fraction to be rerun.
3. Crude solvent naphtha fraction.
4. Heavy naphtha fraction.

The residue left in the still is rerun in the crude straight pipe stills as described in the first distillation of light oil. The crude benzol and toluol fraction is rerun in a vertical still with a steam coil and column, and separated into:

1A. Crude benzol.
1B. Intermediate fraction to be rerun.
1C. Crude toluol.

All is gradually accumulated into fractions 1A, 1C, 3 and 4, which come into commerce as "straw color benzol," "straw color toluol," "crude solvent naphtha," and "heavy naphtha."

Refined Benzols from Crude Fractions

The crude fraction is placed in an iron cone bottom agitator provided with a mechanical stirring device of the propeller type, and there treated with sulphuric acid, the amount varying with the nature of the material. The sulphuric acid polymerizes and removes in the form of tar the unsaturated hydrocarbons, leaving the aromatic hydrocarbons practically untouched. After the sulphuric acid treatment, the acid is carefully drawn off, together with any tarry matter which collects immediately above the acid. The acid treatment is followed by a treatment with dilute caustic soda to remove small amounts of acid left on the sides of the agitator and mixed through the oil. It is then run to a vertical still, provided with steam coil and live steam. In the case of the lower boiling materials, crude benzol and toluol, the distillation can be carried on almost entirely without live steam, but with the higher boiling materials live steam is necessary to complete the distillation. The product of this distillation is refined 90% benzol, from the crude benzol fraction, commercial toluol from the toluol fraction, refined solvent naphtha from the crude solvent naphtha fraction, and 200° naphtha from the heavy naphtha fraction. 50% benzol and 100% benzol are made in a similar way by altering as necessary the distilling points on the crude column distillation of fraction Number 1, as denoted above.

The 50, 90 and 100 per cent benzols consist mainly of benzene and toluene, with small amounts of xylene, and their names are not indicative of the comparative purity of the materials, but merely of the amount distilling at 100° centigrade.

The lower boiling benzols can have the following composition:

	100% Benzol	90% Benzol	50% Benzol
Benzene	94–97%	80–85%	40–50%
Toluene	2–6%	10–20	30–60
Xylene	0–1	0–5	0–25

Pure Hydrocarbons

The manufacture of these consists in taking a refined fraction, such as obtained from the process described in the preceding paragraphs, and submitting it to column distillation in a steam still with a very high column, and taking off a fraction within very narrow temperature limits. Only benzol (benzene) and toluol (toluene) are separated in the pure condition, that is, with less than 0.5% of impurities. A substance called "pure xylol" is manufactured, consisting of a mixture of the three isomeric xylenes. No attempt is made in this country to separate the three isomers. Pure benzol has about the following composition:

Benzene	99.5–99.8%
Toluene	0.05%
Thiophene	0–0.10%
Carbon bisulphide	0–0.02%
Unsaturated hydrocarbons	0
Paraffine hydrocarbons	0–0.55%

Testing of Benzols

For a description of the various tests used on benzols, that is, boiling point, specific gravity, flash, evaporation test, freezing point, and sulphuric acid test, the author has already published these (Journal of Industrial and Engineering Chemistry, Volume 3, No. 1, page 10, January, 1911), and therefore it does not seem necessary to go into these in detail here.

Properties of Benzols.

These can best be given in tabular form, and we present here the average distillation tests, specific gravity, flash, evaporation, sulphuric acid tests, etc., of the refined grades of benzol, and the distillation, specific gravity, flash and evaporation tests of the crude benzols.

Product	Boiling Pt.	Sp. Gr. at 15.5° C.	Flash Point	Evaporation Test
Pure benzol[1]	80–82° C.	.881–.884	Below	10 minutes
100% benzol	100% at 100° C.	.875–.884	Freezing	13½
90% benzol	90% at 100° C.	.875–.882	Freezing	14
50% benzol	50% at 100° C.	.871–.875	Freezing	23
Pure toluol	110–112° C.	.869–.871	Freezing	29
Commercial toluol	90% at 120°	.869–.872	Freezing	33
Pure xylol	135–145°	.865–.867	30° C.	89
Solvent naphtha	90% at 160°	.864–.870	25.5°	107
Straw col. benzol	85–90at 100°	.862–.870	Bel. F.	18
Straw col. toluol	85–90at 120°	.862–.870	Bel. F.	36
Crude solvent	85–90at 160°	.876–.887	25.5° C.	121
Heavy naphtha	85% at 100°	.925–.940	43° C.	303

[1] Pure benzol has a solidifying point of 5.15 to 5.40 C.

Sulphuric Acid Tests: (Water-white grades only)

Pure benzol and toluol, at most pale straw in acid 100%, 90%, 50%, and commercial toluol, yellow in acid, oil white. Solvent, red in acid, oil pale yellow.

Uses of Benzols

In general, the use of all grades of benzol can be summed up in one word, "solvents". They are excellent solvents for gums, resins, greases, rubber, etc., and it is mainly upon the wide and varied solvent power of benzols that the capabilities for their commercial utilization rest. Pure benzol and pure toluol are used as a basis for manufacturing other chemicals, nitrobenzol, aniline, benzoic acid, saccharin, etc., which in their turn are used generally in dye and drug manufacture. Pure benzol and toluol are also used as solvents in cases of continuous extraction where a low boiling solvent of great uniformity is required. Pure benzol is also used for the manufacture of special rubber cements where a very high grade of purity is necessary.

It seems better to classify the uses of the other grades of benzol by the industry in which they are used, rather than by the grade of the material, as the same industry may use a varied number of products for different purposes.

Rubber

Benzol is a perfect solvent for crude Para rubber, being equalled in this respect only by carbon bisulphide. The advantages of benzol over this solvent in regard to safety, etc., are so patent that they need no mention. Therefore, in rubber work where a high grade cement is demanded, benzol is almost universally used. This is particularly the case in the manufacture of automobile tires, and large amounts of benzol are consumed in this industry.

Rubber solutions, too, are used for the manufacture of insulating varnishes, and benzol finds application here also as a solvent. A use in connection with rubber work which has not been very extensively applied as yet in the United States is the cold vulcanization of rubber in which the active agent is sulphur monochloride dissolved in benzol. The use of benzol in this instance depends upon the fact that in addition to being a rubber solvent, it is unaffected chemically by the sulphur monochloride. For most of the rubber purposes, the crude benzols have no application. It is mainly the refined grades that are used, with the lower boiling benzols in greatest use.

Paint and Varnish

The uses of the various grades of benzol in the paint and varnish industry are quite varied, and of late it has been replacing petroleum benzine as a constituent of turpentine substitutes, owing to its greater solvent properties. This is particularly true in certain varnishes where gums, such as Manila, are used, which are not held in solution properly by benzine, but which are perfectly soluble in benzol.

In straight pigment paints, benzol has not found wide application as yet, but it has been found of considerable advantage as a thinner in paints designed for painting resinous woods, owing to the penetration or "tooth" given to the paint. This use has been very well described in a paper by J. Dewar, published in the Proceedings of the convention of Master House Painters and Decorators Association of Pennsylvania, January, 1911, printed in the Painters' Magazine for March, 1911.

Enamel paints, as well as bronze and aluminum paints, also afford a use for benzol. These paints usually have a concentrated solution of Damar gum as their base, and benzol is an ideal material for this purpose, owing to its very strong solvent action on this gum.

Wood stains and stain solvents also have benzol in their make-up. This use is based on the fact that benzols are excellent solvents for oil-soluble aniline colors, and that by the use of benzol, very good penetration into the wood can be obtained.

Paint and varnish removers are to a great extent essentially a mixture of benzol with wood alcohol, and many patents cover the use of benzol in this connection. For these preceding purposes, mostly water white or refined grades are used, and the grade selected depends generally on the evaporative effect desired, in view of the fact that the solvent power of the various grades is approximately the same.

In a few cases in the paint and varnish industry, straw color benzols and the darker, heavier naphthas are used. As examples of this, tar paints, gilsonite paints, ship bottom paints, etc., can be mentioned. In all these the base of the paint is dark, and therefore the color of the solvent used is of no consequence.

Lacquers and Imitation Leather

The writer has classed these two industries together, owing to the fact that the application of benzol is based on the same property in both cases. Benzol by itself is not a solvent for pyroxylin, or soluble cotton. However, if a concentrated solution of pyroxylin in amyl acetate is prepared, this solution can be thinned to a greater extent by benzol than any other commercial solvent, without causing the pyroxylin to be precipitated as a gelatinous, insoluble mass. Numbers of lacquers consist merely of such a concentrated solution of soluble cotton thinned to the utmost degree with benzol. In the imitation leathers in many cases, such a solution is applied to cloth, allowed to dry, and then colored and grained. Besides the pyroxylin imitation leathers, there are also others which have a linseed oil base, and in these also benzol is used for solvent purposes.

Miscellaneous Uses

It hardly seems worth while to the writer to go into any great detail as to the various other uses of benzol much more than a brief mention. Extraction of greases from very varied materials, such as wool, garbage, etc.; extraction of vegetable oils from seeds or press waste; as a constituent of metal polishes, of special metal protective coatings, dry cleaning and scouring; extraction of alkaloids and crude drugs; waterproofing compositions for cloth, etc. As yet it has not been used as a fuel or for motor purposes in the United States, this phase being yet in an experimental stage. In all these uses, the grade of benzol varies according to the particular needs of the process in which it is used.

Production and Distribution of the Use of Benzols, 1910–1911:

In 1910, the production of coal tar solvents of all grades amounted to about 1,500,000 gallons, and in 1911 this was increased to very nearly 2,000,000 gallons. All of this, as noted before, was not produced from coal tar light oil. This was distributed among various industries approximately as follows:

Paint and varnish	47.0%
Rubber and rubber cements	18.0%
Imitation leathers	10.0%
Chemical manufactures	11.0%
Miscellaneous and undetermined	14.0%
	100.0%

Of course, these figures are only approximate, but are indicative of the magnitude of the coal tar solvent industry in this country

Working up Carbolate to Crude Carbolic Acid

The soda solution of tar acids obtained in the working up of light oil serves as raw material for the manufacture of various refined carbolic acids. The carbolate is first boiled to remove mechanically dissolved hydrocarbons. This is, in some cases, performed in an open tank, with coil and live steam; in more improved forms of apparatus, vacuum pans are used for this purpose. From the carbolate thus purified, carbolic acid is liberated with carbon dioxide gas obtained from a furnace burning a mixture of coke and limestone. When the carbolate is sufficiently blown, a clear separation takes place, the upper layer being crude carbolic acid, and the lower, carbonate of soda. The carbonate of soda is treated in another agitator with lime obtained from the carbon dioxide furnace, filter-pressed, and the regained caustic soda used over again in future operations. The crude carbolic acid thus obtained contains from 12 to 16 per cent water, and from 4 to 5 per cent of non-volatile residue, the main amount being phenol and cresols.

Refined Carbolic and Cresylic Acids

The crude carbolic acid is distilled in a vacuum column still, taking off first, water, then a fraction rich in phenol, an intermediate fraction to be rerun, and a cresylic fraction. The cresylic fraction is redistilled in a still using a block tin condenser and receiver, and comes on the market as "white cresylic acid," or, if fractioned to a close boiling point, as U. S. P. cresol.

The first fraction, rich in phenol, is cooled to a suitable temperature, and the phenol crystallizes out, and after draining and centrifugalling, requires only a distillation, using, as in the case of white cresylic, a block tin condenser and receiver, to be marketed as "crystallized carbolic acid," with a melting point of 39 to 40° centigrade. The liquid drainings from the crystal acid are rerun in the column still similarly to the crude acid.

Other Commercial Carbolic Acids

Various other grades of carbolic acid are known as "95% dark", "97–99% straw color," and various so-called "percentage" acids, 15, 25 and 50 per cent, etc., first and second quality. These are obtained from the drained oil from crude naphthalene. The 95% dark is a dark colored carbolic acid, containing at least 95% of bodies soluble in soda, the remainder being made up of water and neutral hydrocarbons.

The 97–99% straw color acid consists merely of the 95% dark distilled under vacuum to remove water and slight amounts of tarry matter, and contains as its only impurity neutral hydrocarbons.

The percentage acids are creosote oil with various guaranteed percentages of tar acids, and are obtained by simple mixture.

The second quality acids deposit naphthalene if chilled, whereas the first quality are free from naphthalene down to at least 32°F.

Methods of Testing Carbolic Acids

The percentage acids are usually tested only for water, percentage of tar acids, and deposit of naphthalene at 32°F. The tar acid test consists in distilling 100 cubic centimeters of material from a bulb and noting the shrinkage of the distillate when treated with 10% caustic soda. Water is determined in the distillation. A small sample is also cooled in a tube to 32° F. for some time to note if naphthalene separates.

97 to 99% straw color acid and 95% dark are merely tested for water, specific gravity, distillation (similar to benzol distillation) and matter insoluble in soda.

Crystal carbolic acid is usually tested by the methods as given in the United States Pharmacopœia, the length to which these tests are carried varying with the purpose for which the acid is to be used.

Uses of Pure Carbolic Acid and Pure Cresol

Pure carbolic acid and pure cresol are used mainly medicinally, and in the manufacture of medicinal preparations. Some carbolic acid is used for the manufacture of picric acid and dyes, and a small amount for leather finishing.

The crude grades of carbolic acids are used in the manufacture of sheep dips and disinfectants, and also in some cases, directly for disinfectant purposes. A small proportion is used in rubber reclaiming and in certain paint and varnish removers. Another interesting use which may be of considerable future importance is in the manufacture of various resinous products by condensation with aldehydes.

Production of Carbolic Acid

It was impossible to obtain reasonably accurate figures for production and importation of refined and crude carbolic acids. The importations are not well enough classified to obtain the definite information desired, nor was the writer able to obtain full information as to that manufactured in this country, especially in the case of the cruder grades.

Naphthalene Manufacture

The crude naphthalene obtained by centrifugalling the second fraction in the first distillation of light oil is melted up and run out into shallow pans and allowed to cool and crystallize slowly. This recrystallized naphthalene is submitted to a second centrifugalling, and is then known as purified naphthalene. A very little goes into the trade as such. For further purification, this naphthalene is distilled and water and tarry matter removed. The middle fraction is run into a lead-lined washbox, and there treated hot with concentrated sulphuric acid, then with water, and finally with soda to remove the last traces of acid. The washed naphthalene is then run back into a still and redistilled, and the middle portion taken as refined naphthalene. The "heads and tails" running from the still are returned to crude naphthalene.

The refined naphthalene is then made into various forms used commercially.

Flake Naphthalene

This is produced by subliming the refined naphthalene from shallow pans into a large wooden room. The naphthalene is held in the pans at about 160°C. by means of steam coils, and gradually sublimes into the room and condenses on the walls, ceiling and floor. After a sufficient period of time has elapsed, the room is opened, the flake naphthalene removed, and a new operation started.

Naphthalene Balls

These are made by molding refined naphthalene in suitable molds.

Other Forms

The flake and balls are the most used forms, but small amounts are put out in the form of crushed crystals, powdered, and various shaped blocks (square, lozenge, domino shape, etc.).

Methods of Testing Naphthalene

Refined naphthalene is tested usually only for melting point. Occasionally a test for coloration with sulphuric acid is made for the very highest grade used for chemical purposes. The most essential features for ordinary purposes is the color, and, moreover, the capability of keeping the white color even in the presence of sunlight. The crude naphthalene are usually assayed by distillation for water and non-volatile residue, in addition to melting point test.

Uses of Naphthalene

The main use of naphthalene is as an insect deterrent. The well-known use of so-called "moth balls" and flake naphthalene requires no comment. A small amount of naphthalene is used in the manufacture of other chemicals, such as naphthol, phthalic acid, etc. However, these latter industries have not as yet been developed to any great extent in the United States.

Production of Naphthalene, 1910–11

In 1910 and 1911, between six and seven million pounds per year were used in the United States, of which approximately 45% was produced in this country. At least 95% is used by the drug trade for sale as a moth deterrent. The remainder is absorbed by manufacturers for divers purposes.

CONCLUSION

The above paragraphs give a general view of the present status of refining light oil in the United States. Besides, at present, some light oil is produced so far from refineries as to make it impractical to transport it to a point where it can be utilized. Such light oil is mostly used up as a thinner in cheap tar paints.

Up to the present, the manufacture of products in this field has been mainly confined to materials already existent in the crude oil, and capable of being isolated by physical and simple chemical means. The further, more intricately chemical manufacture of the various derivatives of benzene, toluene, naphthalene, etc., which serve as raw materials for dye manufacture, has as yet been only carried on in a comparatively small way in the United States.

The chemical possibilities along these lines are yet to be developed in this country, and here indeed is a wide field for technical endeavor which will place the coal tar industry of the United States in a stage of development equal to that already attained abroad.

DIE OBERE BRAUCHBARKEITSGRENZE DES ABEL-PENSKYSCHEN APPARATS UND SEINE VERGLEICHUNG MIT DEM PENSKYSCHEN FLAMMPUNKTPRUFER

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Auf dem Wiener Petroleumkongress ist beschlossen worden, ausschliesslich den Abel-Penskyschen Petroleumprober unter der Bezeichnung als Abel-P-Apparat für alle Staaten zur Bestimmung des Entflammungspunkts von Petroleum anzunehmen. Dabei wurde die Frage berührt, bis zu welchen Temperaturen aufwärts der Apparat benutzbar ist. Nach den deutschen Vorschriften für die zollamtliche Abfertigung von Mineralölen kann er zu Flammpunktsbestimmungen bis 50°C gebraucht werden, wobei das Wasserbad auf 85° C zu erhitzen ist. Für Temperaturen über 50° trat bisher der Penskysche Flammpunktprüfer an die Stelle des Abelapparats, obwohl letzterer zufolge einer mündlichen Mitteilung des Herrn Professor *Holde* in einzelnen Fällen schon bis zu 60 oder 70°C herauf verwendet worden ist.

Um die Unsicherheit über die Verwendbarkeit des Abelapparats in dieser Hinsicht zu beseitigen, wurde auf dem Wiener Kongreb weiter beschlossen, die obere Brauchbarkeitsgrenze des Abelapparats durch eine besondere Untersuchung festzustellen und damit seine Vergleichung mit dem Penskyschen Flammpunktprüfer zu verbinden.

Die hierzu erforderlichen Versuche sind in der Physikalisch-Technischen Reichsanstalt unter meiner Leitung durch die Herren *P. Hebe* und *W. Wuntke* ausgeführt worden. Wir benutzten dazu zwei der Reichsanstalt gehörende Abelsche Petroleumprober (S & R 2902 und 2903) und drie Penskysche Flammpunktprüfer (S & R 2048, 2049, 2050) die von der Firma *Sommer & Runge* in Friedenau-Berlin zur Verfügung gestellt waren. Sämtliche Apparate entsprechen in ihren Abmessungen den amtlichen Vorschriften.

Die den Abelprobern gewöhnlich beigegebenen Thermometer zeigen nur bis 50°C an; um die Apparate für die höheren Temperaturen gebrauchen zu können, wurden sie mit neuen, bis 110° reichenden Thermometern ausgerüstet und dann an die Normal-Abelprober der Reichsanstalt angeschlossen.

Ferner schien es zweckmässig, für die Versuche bei höheren Temperaturen das messingne Heizbad durch ein solches aus Kupfer zu ersetzen, das von gleichen Abmessungen wie jenes ist, aber ein nach unten verlängertes Abflussrohr hat und mit zwei festen hölzernen Handgriffen versehen ist. Es zeigte sich, dass die Apparate im kupfernen Gafäss einen um 0,4°C niedrigeren Entflammungspunkt angeben, was wohl der besseren Leitfähigkeit des Kupfers zuzuschreiben ist.

Für Temperaturen in der Nähe von 100°C und darüber konnte zur Füllung des Heizbades natürlich Wasser nicht mehr verwendet werden, an dessen Stelle Palmin genommen wurde, das Erhitzungen bis 250°C verträgt, ohne sich erheblich zu zersetzen. Ein Einfluss der Palminfüllung auf die Angabe des Entflammungspunkts konnte *nicht* festgestellt werden. Höhere Temperaturen als 230°C kamen bei den vorliegenden Versuchen für das Heizbad des Abelprobers nicht in Betracht.

Die Vergleichung des Abelprobers mit dem Penskyschen Flammpunktprüfer geschah in drei Versuchsreihen, dabei wurden Ole mit Entflammungspunkten von 42, 48, 59, 69 bis 73, 83 und 104°C verwendet. Das Ol mit 42° Entflammungspunkt war russischen Ursprungs, dasjenige mit 48° französischen, während die übrigen Ole due höheren Fraktionen eines amerikanischen Petroleums mit 20, 5° Entflammungspunkt waren.

Bei der ersten Versuchsreihe wurde für die Abelprober das Wasserbad, für die übrigen Reihen das Palminbad benutzt.

Bei den Versuchen im Wasserbehälter wurde die Temperatur des Wassers um 35° höher gehalten als der Entflammungspunkt, nur bei den Olen mit Entflammungspunkt von 70° liess sich dies nicht einrichten, da sonst der Siedepunkt des Wassers überschritten wäre. Wir begnügten uns damit, die Wassertemperatur auf 99° zu halten. Die Versuche bei 70° wurden im Palminbad wiederholt, welches auf höhere Temperaturen gebracht werden konnte.

Der Testanfang, d.h. die Temperatur des Oles, bei der mit dem Eintauchen der Zündflamme in das Petroleumgefäss begonnen wurde, lag bei beiden Apparattypen 5 bis 6° unter dem Entflammungspunkt. Bei den Abelprobern wurde von 0,5 zu 0,5°, bei den Penskyschen Apparaten von 1 zu 1° getestet, beide Male mit zwischentesten von 0,25 auf 0,75°, bezw. von 0,5 auf 1,5°.

TABELLE 1

Verleichung des Abelprobers mit dem Penskyschen Flammpunktsprufer bei 42°, 48°, 50°, 69° bis 73°, 84°, 104°.

für beide Apparattypen		Abelprober mit kupfernem Erhitzungsgefäss			Flammpunktsprüfer		Unterschiedrivischen den korrigierten Angaben der Abelprober und der Flammpunktsprüfer
Heigerung der Temperatur des Oles in 1 Minute	Testanfang	Temperatur des Wasserbades (1. Reihee), berio, des Palminblades (2. und 3. Reihe)	Mittlere Temperatur des herausragenden Quecksilberfadens	Entflammungspunkt; korrigiertes Mittel aus den Angaben von 2 Apparaten	Temperatur des herausragenden Quecksilberfadens	Entflammungspunkt; korrigiertes Mittel aus den Augaben von 3 Apparaten	
		1. Versuchsreihe (März-April 1912)					
bei Abelprober I,6° C bei Flammpunktsprüfer 2°	35 C	78 C	28,3 C	42,1 C	31,7 C	41,9 C	+0,2°C
1,5 C	42	85	28,3	28,3	48,2	33,1	+0,5
1	54	95	28,2	59,3	36,5	58,7	+0,6
0,67	64	99	24,3	24,3	70,0	69,3	+0,7
		2. Versuchsreihe (April 1912)					
0,67	65	125	——	73,0	——	72,2	+0,8
1	65	130	31,2	72,3	35,7	71,6	+0,7
3	63	165	34,7	70,1	38,6	69,4	+0,7
6	63	220	45 7	68,6	43,6	69,4	—0,8
3	75	195	41,7	83,1	39,2	83,5	—0,4
3	77	195	——	83,0	——	83,9	—0,9
3	96	230	40,8	104,0	44,2	103,5	+0,5
		3. Versuchsreihe (Mai 1912).					
3	34	120	28,5	40,5	30,3	40,0	+0,5
3	43	135	32 4	49,4	34,2	49,4	0,0
3	53	150	32,7	58,7	35,7	59,3	—0,6
3	65	165	32,9	70,8	70,8	71,1	—0,3
3	78	185	30,1	83,6	36,9	83,9	—0,3
3	98	220	34,6	103,9	43,2	103,4	+0,5
						Gesamtmittel	+0,1° C

Die Temperatursteigerung des Oles schwankte bei der ersten Versuchsreihe zwischen 0.67 und 2° in der Minute, bei der zweiten Reihe zwischen 0,67 und 6° in der Minute, während bei der dritten Reihe die Steigerung in allen Fällen 3° in der Minute betrug.

Bei fast allen Versuchen ist die mittlere Temperatur des herausragenden Quecksilberfadens beobachtet und die daraus folgende Korrektion an die Thermometerablesungen angebracht worden. Diese Korrektionen sind in dem Falle nicht zu vernachlässigen, wenn das Thermometer mit ganz eintauchendem Faden geprüft ist, da sie in den höchsten Temperaturen (bei 104°) bei den Abelprobern bis 1,2° und bei den Flammpunktprobern bis 0,7° ansteigt.

Die Resultate der drei Versuchsreihen sind in der folgenden Tabelle 1 zusammengestellt; hierbei sind die Zahlen jeder Horizontalreihe die Mittel aus mindestens sechs einzelnen Vergleichsreihen.

Die Zahlen der letzten Vertikalkolumne geben die Unterschiede zwischen den korrigierten Angaben der Abelprober und der Penskyschen Flammpunktprüfer für die Temperaturen zwischen 40 und 104° C. Das Gesamtmittel beträgt +0,1°, von welchem die einzelnen Zahlen um rund ± 1° abweichen. Die beiden Apparattypen zeigen also innerhalb dieses Temperaturintervalls selbst bei einer Verschiedenheit der Temperatursteigerung im Olgefäss von $\frac{2}{3}$ bis 6° in der Minute bis auf ± 1° C übereinstimmende Entflammungspunkte.

Aus den hier nicht angegebenen Abweichungen der Angaben der einzelnen Prober von den Mitteln berechnet sich der mittlere Fehler eines aus sechs einzelnen Vergleichungen gebildeten Mittels für die Abelprober zu ± 0,11° und für die Flammpunktprüfer zu ± 0,23°, für letztere also doppelt so gross wie für erstere, was zu erwarten war, da beim Testen das Eintauchen der Zündflamme in das Olgefäss beim Flammpunktprüfer in doppelt so grossen Temperaturabständen geschieht wie beim Abelprober.

Aus den Zahlen der Tabelle geht unzweifelhaft hervor, dass der Abelsche Apparat in der oben angegebenen Modifikation unbedenklich zu Flammpunktsbestimmungen mindestens bis 104° C benutzt werden kann. Dies ist wichtig, da für die neuen Vorschriften des Jnternationalen Ubereinakommens über den Eisenbahnfrachtverkehr bezüglich der Beförderung von Petroleum und anderen Olen vorgeschlagen ist, dass diese unter den Bedingungen für brennbare Flüssigkeiten zugelassen werden sollen, wenn sie im Abelapparat nicht unter 21°C und im Penskyschen Flammpunktprüfer nicht oberhalb 100° C entflammen. Man wird also für diese Zwecke künftig mit dem Abel-P.-Apparat allein auskommen.

Ausserdem übt die Temperatursteigerung beim Flammpunktprüfer einen grösseren Einfluss auf den Entflammungspunkt aus als beim Abelprober, wie aus den in nachfolgender Tabelle 2 zusammengestellten Versuchen hervorgeht.

TABELLE 2

Einfluss der Temperatursteigerung auf den Entflammungspunkt.

Abelprober		Penskyscher Flammpunktsprüfer			
Temperatursteigerung in 1 Minute	Entflammungspunkt Öl I	Temperatur steigerung in 1 Minute	Entflammungspunkt		
			Öl II	Öl II	Öl IV
1° C	43 65° C	2° C	49,3° C	77,9° C	101,0° C
2,5	43,4	5	47,8	77,1	100,4
		8	47,0	76,0	100,2

Der Entflammungspunkt wird also bei schnellerer Temperatursteigerung im allgemeinen erniedrigt; bei den Abelprobern aber nur um $\frac{1}{4}$°, bei den Flammpunktprüfern dagegen um mehr als einen Grad. Sobald es sich also bei der Bestimmung des Flammpunkts um eine grössere Genauigkeit als 1° handelt, empfiehlt es sich, den Abelapparat in der angegebenen Modifikation zu verwenden.

NEUERUNGEN AUF DEM GEBIETE DER TORFGEWINNUNG UND TORFVERWERTUNG

Von Dr. W. Wielandt
Oldenburg i/Gr.

Seitdem die Gestehungskosten für Steinkohle in allen Ländern erheblich gestiegen sind, haben die geringeren Brennstoffe wie Braunkohle, Torf und dergl. erhöhte Beachtung gefunden, und insbesondere hat der Torf die Aufmerksamkeit auf sich gezogen, weil er gerade in kohlearmen Ländern am reichlichsten vorhanden ist. Eine grosse Torfindustrie hat sich indessen bisher noch nicht recht entwickeln können, weil eine solche einerseits mit reinem Handbetrieb selbstverständlich undenkbar war, anderseits die bisher verwandten Torfmaschinen eine Unmasse von geschulten Arbeitern erforderten, sodass der Torf nur noch teurer wurde als mit Handbetrieb.

Die letzten Jahre haben hierin nun eine entscheidende Neuerung gebracht, nämlich die Torfmaschinen des Dr. Wielandt in Oldenburg, der seit Jahren sich ausschliesslich der Torfmoorverwertung widmet und das von ihm 1905 gegründete Torfverwertungsunternehmen zu Elisabethfehn im Grossherzogtum Oldenburg jetzt zum grössten torfindustriellen Betriebe in Oldenburg ausgedehnt hat.

Die neuen Torfbagger mit dem angefügten Torfsodenausbreiter arbeiten im Gegensatze zu den bisher gebräuchlichen Torfmaschinen vollständig automatisch, indem sämtliche Arbeitsvorgänge, nämlich das Baggern der Moormasse, das Mischen und Formen derselben, das Abteilen in einzelne Soden, das Ausbreiten der Torfsoden auf dem Trockenfelde, sowie das Nachbunken, von der Maschine selbsttätig ausgeführt werden, sodass lediglich für die Beaufsichtigung, für die Bedienung der Motoren und das Legen der Geleise je nach den Moorverhältnissen 2–3 Arbeiter gebraucht werden. Der Torfbagger besteht aus einem auf Feldbahngeleisen on 600 mm Spur oder auf einzelnen Schienenstücken sich mit einer Stundengeschwindigkeit von 20–30 Meter gleichmässig vorwärtsbewegenden Fahrgestell, einer seitlich auf demselben befestigten, 2 bis 5 Meter tief und 0,8 bis 1,2 Meter breit arbeitenden Eimerkette, einem doppelschneckigen Mischwerke, in welches die Moormasse aus den Eimern vermittelst eines Schüttrichters gelangt, einem Pressenmundstück von beliebigem meist fünfeckigem Querschnitte, einem Sodenabschneider, der den Torfstrang automatisch in gleich langen Soden zerteilt und einer Abbunkvorrichtung, welche hinter der Baggerleiter die Bunkerde abfgräbt und seitlich in den Püttengraben abwirft. An das Maschinengestell angeschlossen und mit ihm kontinuierlich sich vorwärtsbewegend ist der Torfsodenausbreiter, welcher vermittelst eines Bandes, dessen einzelne Tragplatten an einem endlosen Zugseil drehbar befestigt sind, die Torfsoden vom Mundstück aufnimmt und sie, sobald das Band bis zum Ende vollgelaufen ist, gleichzeitig auf dem etwa 15 bis 30 Meter breit zu nehmenden Trockenfelde in horizontaler Lage abwirft. Der Antrieb der Maschine erfolgt durch Elektromotor oder durch einen Benzinmotor, der entweder auf dem Maschinengestell selbst oder auf einem besonderen Fahrgestell angebracht ist. Die Bagger werden hergestellt für eine Arbeitsleistung von 60 bis 70 Kubikmeter Rohmoor stündlich entsprechend 5 bis 10 tons lufttrockenem Torf, wiegen komplet mit Sodenausbreiter je nach Grösse 3000 bis 5000 Kilogramm und bedürfen etwa 20 bis 32 PS.

Neuartig an der Betriebsweise dieser Maschinen ist zunächst, dass nur Streifen von durchschnittlich 1 Meter Breite abgetorft und ein Trockenfeld von durchschnittlich 20 Meter Breite genommen wird, während bisher Streifen von 3 bis 4 Meter Breite abgetorft und auf 80 bis 100 Meter Breite ausgelegt wurden. Nach dieser neuen Methode gelangt stets nur die abgetrocknete Moorwand zur Verarbeitung, welche etwa doppelt so viel Trockensubstanz im Kubimeter enthält wie die weiter im Moore liegenden Schichten, wodurch die Torfsoden von vornherein wesentlich trockener sind als bei der alten Methode. Damit wird erreicht dass die Torfsoden je nach der Witterung bereits nach 4 bis 10 Tagen gehäufelt und nach insgesamt 2 bis 4 Wochen vom Trockenfelde abgenommen und in grosse Haufen aufgestapelt werden können. Die Folge hiervon ist, dass das selbe Trockenfeld in einem Sommer wiederholt befahren und belegt werden kann und dass die Torfkampagne wesentlich länger in den Herbst ausgedeent und so Maschinerie und Moor viel besser ausgenutzt werden kann als bisher. Damit ist ein wesentlicher Schritt zu dem Ziele der Torfschnelltrocknung getan, ohne dass es einer künstlichen Trocknung durch Wärme, einer Wasserauspressung oder sonstigen kostspieligen Behandlung bedürfte. Ausserdem aber stellen sich die Selbstkosten für die Trocknung und das Einsammeln des Torfes geringer als bisher, weil die Soden nicht über eine so ausserordentlich breite Fläche ausgelegt werden, sondern nur auf einer schmalen Fläche liegen und dadurch dem Abfuhrgeleise bedeutend näher sind. Neuartig ist ferner an dieser Maschine, dass schräge Abhänge abgetorft werden, deren Neigungswinkel je nach der Weichheit des Moores eingestellt werden können. Dadurch wird offensichtlich eine grössere Standfestigkeit der Anhänge gegen den Seitenschub der Moormasse erreicht, und ermöglicht, dass auch in schlecht entwässerten Mooren gearbeitet werden kann, sodass man unter Umständen mit der Abtorfung eines frischen Moores um 2–3 Jahre früher beginnen kann als dies bisher der Fall war. Dies wird ganz wesentlich auch dadurch unterstützt, dass die Maschine ein ganz geringes Gewicht hat und infolge des gleichmässigen Ganges dem Moorboden keinerlei Erschütterungen mitteilt.

Die weiteren Vorzüge des neuen Systemes sind, abgesehen von der Reduzierung der Bedienungsmannschaften auf ein Sechstel der bisherigen, noch folgende.

1. Sämtliche Bewegungen sind kontinuierlich und durchaus gleichmässig, wodurch Konstruktion und Betrieb sehr einfach, der Kraftbedarf gleichbleibend und die Abnützung sehr geringist.

2. Die Baggerleiter kann während des Betriebes ohne Stillsetzen der Maschine gehoben und gesenkt und ebenso schräger oder steiler eingestellt werden, wodurch Neigungswinkel und Tiefgang augenblicklich der ja oft wechselnden Moortiefe und Moortrockenheit angepasst, und eventuelle Hindernisse wie Sandhügel und Baumstämme ohne Zeitverlust umgangen werden können.

3. Bei der Ueberführung der Maschine von einem Moore zum anderen wird die Baggerleiter hochgezogen und quergestellt,

sodass die Maschine jedwede Wendung und Platzveränderung ohne irgend welche Demontage in wenigen Stunden bewerkstelligen kann, wozu bei den alten Systemen mehrere Tage notwendig sind. Der Transport des Baggers selbst auf grosse Entfernungen ist sehr bequem, da derselbe auf normalen Feldbahngeleisen läuft.

4. Da die Baggerbecher gleichzeitig alle Schichten des Moores abbaggern, erfolgt bereits in den Bechern eine Mischung der verschiedenen Schichten, und es resultiert ein wesentlich gleichmässigeres und dichteres Produkt als beim Einwerfen der Moormasse mit Hand wie früher.

5. Infolge des geringen Arbeiterbedarfes kann auch nachts bei elektrischer Beleuchtung gebaggert werden, sodass Maschine und Moor fast doppelt so lang wie bisher ausgenützt werden können.

6. Der Bagger kann auch in holzreichen Mooren arbeiten, da die Baggerungsbreite nur gering und der Püttengraben von aussen sehr gut zugänglich ist, sodass etwa vorhandene Wurzeln ohne grosse Schwierigkeit mit Hand von der Seite her entfernt werden können, bevor die Becher an sie anstossen.

Die Torfgewinnungskosten mit diesen Baggern stellen sich je nach den speziellen Moorverhältnissen durchschnittlich auf etwa *M* 4.—bis *M* 5.—pro 10000 *kg* lufttrockenen Torffür das Baggern einschliesslich des Kraftverbrauches, *M* 7.—für das Trocknen, *M* 4.—für Materialien, Reparaturen und Aufsicht, *M* 4.—bis *M* 5.—für den Transport bis zur Verwendungsstelle und *M* 4.—bis *M* 5.—für Abschreibungen auf Maschinen, Geleise und Wagen, insgesamt also auf durchschnittlich etwa *M* 25.—pro 10000 *kg* frei Verwendungsstelle.

Mit diesen neuen Maschinen sind nun hervorragende Resultaten erzielt worden auf dem vorhin genannten Torfwerke zu Elisabethfehn, wo bereits vier verschiedene Exemplare in Dauerbetrieb stehen. Diese Bagger haben dort im vergangenen Sommer zum Teil bis zum 16. September gearbeitet, während die alten Maschinen in der dortigen Gegend sämtlich schon in den ersten Julitagen stillgesetzt werden mussten, was also eine Verlängerung der Kampagne auf nahezu das Doppelte bedeutet. Ferner wird dort die häufig wiederholte Bearbeitung und Belegung desselben Moores in einem Sommer durchgeführt, indem z.B. eine jener Maschinen im gegenwärtigen Sommer regelmässig aller zwei bis drei Wochen auf dem gleichen Moore arbeitet und den Torf ausbreitet, mithin dasselbe Moor etwa acht bis zehnmal in einem Sommer bearbeitet, während sich nach den bisherigen Trocknungsmethoden auf demselben Moore stets nur ein einmaliges Belegen des Trockenfeldes als möglich erwiesen hatte. Auch hat sich in Elisabethfehn in der Tat gezeigt, dass der mit den neuen Baggern erzeugte Torf infolge der tadellosen maschinellen Bearbeitung derart fest und dicht wird, dass er beim Durchsägen eine gleiche Strucktur aufweist wie hartgepresste Briketts oder künstliches Holz.—Im übrigen werden auf dem Moore zu Elisabethfehn die Bagger mit Benzinlokomobilen nach Entwürfen von Dr. Wielandt angetrieben, die nur 800 bis 1200 *kg* wiegen und im Herbste auch zur Anlieferung des Torfes verwandt werden können.

Aus Vorstehendem ergiebt sich, dass mit dieser neuen Torfmaschine der Weg gebahnt ist zu einer grossen und rationellen Torfgewinnungsindustrie.

Nun sind bisher die Ansichten noch sehr geteilt über die Frage, in welcher Weise grosse Torfmengen am besten zu verwerten sind. Zu diser Frage sei mitgeteilt, dass seit 1907 zu Elisabethfehn in Oldenburg eine Torfverkokungsanlage System Dr. Wielandt arbeitet, welche in fünfjährigem Bestehen den Beweis erbracht hat, dass Torfverkokung nach diesem System äusserst gewinnbringend ist und ganz zweifellos für die Verwertung grösster Torfmengen sehr geeignet ist.

Die genannte Anlage besteht aus drei Verkokungsöfen mit einer Kapazität zur Erzeugung von rund 9 tons Torfkoks pro 24 Stunden aus rund 30 tons Torf, den dazugehörigen Anlagen für elektrische Torfförderung und Koksstapelung, einer Torfteerdestillation zur Aufarbeitung des Torfteers zu Kreosotöl, Gasöl, Paraffin und Pech und einer Schweelwasserdestillationsanlage zur Gewinnung von essigsaurem Kalk und Schwe felsaurem Ammoniak. Das Prinzip des vielfach patentierten Verkokungsofens beruht darauf dass durch die Anbringung einer Verbindungsrohrleitung zwischen dem obersten Ofenteil und dem unteren Kokskühlraume und die gleichzeitige Verlegung des Gasabsaugepunktes etwa in die Mitte des Ofens eine Verbindung zwischen der Wasserentwickelungszone und der Gasentwickelungszone geschaffen ist. Durch diese Verbindungsleitung werden die zahlreichen bei der Trocknung des Torfes im oberen Ofenteil entstehenden Wasserdämpfe allein infolge der natürlichen Druckverhältnisse in dem Kokssammeltrichter gesogen, durchstreichen den mittleren Ofenteil, in welchem die permanenten Gase und die Teerdämpfe entwickelt werden und verlassen mit diesen zusammen den Ofen. Die wertvolle Folge dieser Anordnung ist, dass der glühende Koks bereits innerhalb der Retorte automatisch abgelöscht wird, sodass er den Ofen fast kalt verlässt, dass durch die Einwirkung des Wasserdämpfe auf dem entgasenden Torf wesentlich mehr Ammoniak gewonnen und die Teerdämpfe vor zu weit gehender Zersetzung bewahrt werden und dass die Destillationstemperatur erniedrigt wird. Ein weiterer technischer Vorteil ist, dass sowohl ganz oben als auch ganz unten im Ofen nur Wasserdämpfe herrschen, sodass beim Einfüllen des Torfes und Entleeren des Kokses keine wertvollen Heizgase, sondern nur überflüssiger Wasserdampf verloren geht und ausserdem eine Feuersgefahr ausgeschlossen ist. Schliesslich giebt dies Prinzip auch eine sehr wertvolle automatische Regulierung des Ofenganges, indem gerade dann, wenn einmal übermässig feuchter Torf in die Retorten kommt, aus dessen Wasser in Berührung mit dem Koks eine Vermehrte Menge Wassergas gebildet wird und durch die solcher Gestalt ohne Weiteres vermehrte Gasmenge eine stärkere Beheizung der Retorten gerade dann von selbst erfolgt, wenn diese einer grösseren Wärme zwecks Verdampfung der grössern Wassermengen bedürfen. Dadurch ist es möglich, auch bei feuchterem Torf mit dem eigenen Torfgase nicht nur die Beheizung der Oefen sowie der Nebenbetriebe wie Teerdestillation und Eindampfung der Salze auszukommen, sondern auch elektrische Kraft kostenfrei abzugeben für die Torfmaschinen auf dem Moore.

Von den Produkten wird der Torfkoks speziell für Kupferschmiedezwecke, Qualitätseisenzwecke, Holzkohlenhochöfen und weil dergl. an Stelle von Holzkohle verwendet und dieser vorgezogen, er dichter, härter und ergiebiger ist als jene. Sien Schwefelgehalt stellt sich durchschnittlich auf 0.15 per cent. sein Aschengehalt auf 3 bis 5 per cent. seine Heizkraft auf 7,000 Kalorein pro Kilogramm. Von den Nebenprodukten ist das Gasöl zum Karborieren von Wassergas und zur Oelgasbereitung vorzüglich geeignet, ebenso in gereinigtem Zustande auch als Dieselmotorentreiböl, da es infolge verhältnissmässig hohen Wasserstoffgehaltes eine günstige Entzündungstemperatur hat. Das Pech wird für Kohlenbrikettierungs—und Dachpappenzwecke verkauft.

Die Gestehungskosten für den Torfkoks stellen sich gemäss den Resultaten des beschriebenen Torfverkokungsbetriebes sehr niedrig, nämlich z.B. bei einer Anlage zur Erzeugung von 12,000 tons Torfkoks jährlich einschliesslich der Amortisation der Anlage, abzüglich des Erlöses aus den Nebenprodukte auf etwa *M* 18.—bis *M* 20.—per ton Koks, bei einer Anlage von 30,000 tons Torfkoks jährlich auf weniger als *M* 10.—per ton. Es ist also bei einer derat grossen Anlage sehr gut möglich den Torfkoks mit grossem Gewinn als gewöhnliches Heizmaterial zu verwenden, wofür er sich besonders gut eignet, da es absolut rauch und geruchlos brennt, einessehr geringen Schornsteinzuges bedarf, nur wenig durchaus schlackenfreie Asche, die noch dazu als Düngemittel verwendbar ist, hinterlässt und sehr rasch und intensiv anbrennt.

Als Anlagekapital für eine Anlage zur Erzeugung von 12,000 tons Torfkoks jährlich sind einschliesslich der gesamten Torfmaschinerin, ausschliesslich der Grundstücke durchschnittlich etwa *M* 700.000.—zu rechnen, für eine Anlage von 50,000 tons jährlich etwa *M* 1,500,000.

Es unterliegt keinem Zweifel, dass das beschriebene Torfverwertungsverfahren auch für Amerika von grosser Bedeutung und geeignet ist, die Hebung der in den grossen Moorlagern ruhenden ungeheuren Naturschätze zu fördern.

THE BRIQUETTING OF AMERICAN LIGNITES

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INTRODUCTORY STATEMENT

The industrial supremacy of the United States has depended in no small degree on the extent of its fuels resources and their availability at prices low in comparison with the price of similar fuel in foreign countries. A small portion of the total fuel resources of the United States has been used at the present time, but the increase in the cost of mining, due to increased wages and other reasons, will in time lead to a better utilization of all the waste products from the mining industry, and a development of the types of fuel which are inferior to those now in use and at present not considered practical for fuel purposes.

Of the five general classes of fuel, namely, anthracite, bituminous, sub-bituminous (also called black lignite), lignite (or brown coal), and peat, the anthracite and bituminous are in general use today in the United States, and the sub-bituminous is being used in the West to an increasing extent, but the lignite and peat are at the present time utilized only as local fuels and in a few sections.

In the western and southern parts of the United States there are large deposits of a low-grade fuel called lignite, a form of brown coal. This lignite in its natural condition is not suitable for general use as a fuel mainly for three reasons. First, the high proportion of water or moisture in it (which in some cases is as high as 42 per cent), decreases the heat value of the raw lignite to such an extent that it makes a very inefficient fuel to transport or to use under a boiler. Second, on exposure to the air for a few days, the lignite crumbles or slacks. The lumps check and fall into small irregular pieces that exhibit a decided tendency to break into small thin plates. Hence the lignite deteriorates greatly during storage or long transportation. Third, if large quantities of lignite are stored in piles, spontaneous combustion takes place much more readily with this fuel than with bituminous coal. A peculiar property which was noticed with a sample of lignite that came under the observation of the author was that spontaneous combustion originated in a small pile of dried lignite of only three or four tons in amount, which was in a refuse pile, exposed to sun and rain. Apparently the moisture penetrating to the center of this mass of dried lignite increased its tendency to heat.

Because of its inferiority as a fuel, lignite has not up to the present time found extensive use in this country. It has, to be sure, been used in Texas, North Dakota, and other States to some extent for a local household fuel, and is rapidly gaining favor as a gas producer fuel in the State of Texas. On the other hand in many localities in which lignite is found in abundance, good bituminous fuel is sold at a comparatively high price and for this reason considerable interest has recently been shown in the briquetting of this lignite as a means of improving its heat value and making it available for a household fuel.

German lignite contains a higher percentage of water than the American lignite, the water content of the freshly mined fuel being from 40 to 60. German lignite is usually so soft that it can be cut with a spade. Many lignite beds in Germany are filled with logs and pieces of wood so well preserved in the matrix of partially carbonized material that they burn readily and form a cheap and abundant fuel for steam and other heating at the briquetting plants.

EXTENT OF LIGNITE RESOURCES IN UNITED STATES

Mr. Marius R. Campbell of the United States Geological Survey estimates in 1907 the extent of fuel deposits of the United States to be as follows:[1]

TONNAGE (SHORT TONS) BY GRADES OF COAL AND ACCESSIBILITY

Kind of Coal	Area in sq. miles	Original Supply		Easily accessible coal still available
		Amount easily accessible	Amount accessible with difficulty	
Anthracite and bituminous	250,531	1,257,766,000,000	505,730,000,000	1,247,672,000,000
Sub-bituminous	97,636	356,707,000,000	293,450,000,000	356,594,000,000
Lignite	148,609	389,545,000,000	354,045,000,000	389,534,000,000
Total	496,776	2,004,018,000,000	1,153,225,000,000	1,993,800,000,000

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[1] Mineral Resources, 1907.

A recent estimate made by the Geological Survey at the request of the writer, of the lignite resources of the various states follows, and it will be noticed that both the area of the lignite deposits and their tonnage are each greater than was supposed in the earlier estimate given above:

State	Area in Square Miles		Estimated original lignite supply in short tons both easily accessible and accessible with difficulty
	Containing Workable Beds	May Contain Workable Beds	
Alabama......	*a*	6000	*a*
Arkansas......	100	5900	90,000,000
Louisiana......	8800	*a*	*a*
Mississippi.....	7500	*a*	*a*
Montana......	33,130		365,474,700,000
North Dakota..	29630	6350	697,929,400,000
South Dakota..	2160	820	1,020,300,000
Tennessee.....	1000	*a*	*a*
Texas.........	2000	53000	23,000,000,000
California.....	*a*	*a*	*a*
	156,390		1,087,514,400,000

a Data available is insufficient to base an estimate on.

There are therefore over 156,390 square miles of land in the United States underlaid with lignite of greater or less thickness, and the estimated original supply of lignite both easily accessible and accessible with difficulty was over a thousand billion short tons, of which only a very small fraction of one per cent has been used up to the present time. It will therefore be seen that there is a sufficient supply of lignite for many years.

There are many deposits of lignite coal in the western and southern States, the most important deposits being found in Texas, North Dakota, Montana, and California. These deposits vary in thickness from a few inches up to eighty feet. Lignite is a non-coking fuel, and, therefore, cannot be made into coke in its natural condition, but goes to pieces in a fire, breaking into fine particles the size of coarse sand and in this condition it tends to sift through the grate of a stove. Therefore, special furnaces are required to burn the raw lignite properly.

PRODUCTION AND VALUE OF LIGNITE IN UNITED STATES

In the following table is shown the production and spot value at the mine of lignite fuel mined in the United States in 1910.[1]

Field	Production in 1910 tons	Average price per ton at mine 1910
California	11,164	$1.64
North Dakota	595,139	1.49
Oregon	67,533	3.48
Texas	881,232	.87

BRIQUETTING OF LIGNITE

The fact that Germany and other European countries have carried on the briquetting of lignite for thirty years with success leads one to wonder why the United States has not done more towards developing this material as a source of supply. The main reasons why this has not been done are: first, the low price of high-class American fuels compared with the price of similar fuels in foreign countries. Second, until recently there has been only a small demand for fuel in the Western part of this country in which the larger deposits of lignite are found.

BRIQUETTING TESTS OF UNITED STATES BUREAU OF MINES

In connection with its work of testing fuels, the United States Technologic Branch of the United States Geological Survey, which Branch has been made the Bureau of Mines by an Act of Congress, investigated the briquetting qualities of seven samples of American lignite without binding material with a machine and equipment imported from Germany for this purpose, this outfit being typical of that used in Germany for the production of brown coal briquets. These experiments were carried on at the Pittsburgh plant of the Bureau. This German lignite briquetting equipment was furnished by the Maschinenfabrik Buchau Actien Gesellschaft zu Magdeburg, Germany. As received from the maker, the plant included the following equipment: a Schulz tubular drier, a sorting sieve, auxiliary crushing rolls, a plate type cooler, a screw conveyer, a large storage hopper, a briquetting press directly connected to steam engine, apparatus for conveying the briquets, shafting, hangers, and extra sets of dies for the press. To complete the plant, the Survey erected a coal elevator, crushing rolls, and the necessary belting and piping in addition to the buildings and stacks. The building used to house the briquetting equipment had a steel frame enclosed with curtain walls of reinforced concrete two inches thick, and concrete floors. It was, therefore, very nearly fire-proof.

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[1] Compiled from data in "Production of Coal in 1910" by E. W. Parker. Advance Chapter from Mineral Resources. United States Geological Survey.

A description of the equipment and tests may be found in Bulletin 14, United States Bureau of Mines, entitled "Briquetting Test of Lignite at Pittsburgh" by the author of this article and, therefore, details need not be included here.

This series of investigations was made on the German machine without the use of any artificial binder, the natural inherent bitumen of the lignites yielding the binding material. Therefore those lignites which were not made into good briquets in this series of experiments may possibly be made into excellent briquets in a machine adapted to working with artificial binders. Certainly there is opportunity for further experiment in this direction, and it is to be hoped that the Bureau of Mines may be able to conduct tests of lignite with various binding materials on other types of presses than the German one used on the tests mentioned.

A brief description of the German briquetting press and the method of applying pressure should be included here. This lignite press which as mentioned above is typical of those used in Germany for briquetting lignite without binders, is of the open mould type and pressure is developed by forcing lignite through a gradually decreasing opening or tube three feet long. This press was built to give a working pressure of from 14,000 to 28,000 pounds per square inch and as the area of the surface of the briquet was approximately 15.5 square inches, the total pressure developed by the machine was 217,700 to 435,400 pounds. The capacity of the press was about 3.8 tons per hour, but varied with different lignites according to the ease with which they could be briquetted, this factor governing the speed at which the press could be run. The speed varied from 60 to 120 revolutions a minute, the latter figure being the rated speed of the machine when working on German lignites.

All lignite was shipped from the mines to the plant in box cars to eliminate as far as possible any weathering of the fuel on the road. At the plant, the lignite was stored in covered bins and a quantity sufficient for a test (three or four tons) was prepared for pressing by elevating, crushing, drying and cooling.

ANALYSES OF AMERICAN LIGNITES TESTED BY THE UNITED STATES BUREAU OF MINES

In the following table are shown the localities from which samples of lignite have been obtained for briquetting tests and their proximate analyses as taken from the mine.

PROXIMATE ANALYSES AND HEAT VALUE OF LIGNITES

Field No.	P-7	P-8	P-9	P-11	P-13	P-15	P-14
State	Texas	Texas	Texas	North Dakota	North Dakota	North Dakota	California
County	Milam	Medina	Robertson	Bowman	Stark	Ward	Amador
Locality	Rockdale	Lytle	Calvert	Scranton	Lehigh mine	Vanderwater	Ione
Laboratory No . .	7,270	7,330	7,403	7,499	7,537	7,587	7,621
Air drying loss. . .	31.50	24.20	29.30	36.00	37.80	28.90	32.30
Moisture.	35.30	32.92	34.33	41.43	42.04	36.64	39.46
Volatile matter. . .	26.22	27.42	25.94	23.86	23.40	22.64	29.50
Fixed carbon. . . .	29.58	27.08	30.93	28.45	27.67	30.74	17.55
Ash.	8.90	12.58	8.80	6.26	6.89	9.98	13.49
Sulphur.	.76	1.46	.95	.74	.68	.45	.97
Heat value B.t.u.	6,898	6,840	7,214	6,241	6,079	6,394	6,080
CS_2 Extract (Dry sample). .	1.20	2.01	1.24	1.72	1.43	1.08	7.60

RESULTS OF LIGNITE BRIQUETTING TESTS MADE BY UNITED STATES BUREAU OF MINES

As a result of the tests made by the Bureau of Mines, it was found that briquets could be made by the German method without binder of lignite from the following localities:

Lytle, Medina Co., Texas. Satisfactory briquets made, but difficult to briquet.

Scranton, Bowman Co., N. D. Satisfactory briquets made.

Lehigh, Stark Co., N. D. Difficult to briquet but some fair briquets were made from it.

Ione, Amador Co., California. Excellent briquets easily made.

Lignites from the following localities were not briquetted without binder on the German press by the tests of the Bureau of Mines, but in some cases the amount of the sample was too small to determine with accuracy whether or not the lignite could be briquetted by this method.

Rockdale, Texas. Sample insufficient.

Calvert, Texas. No satisfactory briquets made.

Vanderwalker, N. D. No satisfactory briquets made.

Since making the tests described above on the German press without binder, tests have been made on the hydraulic hand laboratory press of the Bureau, using various binders with some of the lignites which were not briquetted without binder as easily as they should be to be practical, and the indication of the laboratory tests are that the following lignites can be made into satisfactory briquets with the per cent of binder given as the minimum amount that will make briquets for practical purposes.

Sample from	Binder Amount	Kind
Vanderwalker, N. D.	6%	Water gas pitch
Vanderwalker, N. D.	3%	Flour[1]
Vanderwalker, N. D.	8%	Cell pitch not sufficient
Calvert, Texas	5%	Water gas pitch
Calvert, Texas	2%	Flour[1]
Calvert, Texas	8%	Cell pitch[1]

The results of these laboratory tests require checking on a larger scale before it is possible to be entirely sure that the percentages of binders used would be sufficient to make satisfactory briquets.

[1] Of course briquets were not water-proof.

RESULTS OF UNITED STATES BUREAU OF MINES LIGNITE BRIQUETTING TESTS

Heating Values

The following table shows how the heating value of lignite is increased by the removal of moisture during briquetting.

Improvement of heat by briquetting.

Source	Field Designation	Moisture			Heat Value per Pound		
		In Raw Lignite	In Briquets	Removed	Raw Lignite	Briquets	Increase
		Per cent	Per cent	Per cent	B.t.u.	B.t.u.	Per cent
Texas......	Pitts. No. 11	33.0	9.0	24.0	6,840	9,336	36.5
N. Dakota..	Pitts No. 8	40.0	12.0	28.0	6,241	9,354	50.0
N. Dakota..	Pitts. No. 13	42.0	10.0	32.0	6,079	9,355	54.0
California...	Pitts. No. 14	40.0	10.0	30.0	6,080	9,264	52.4

Of the four samples of raw lignite named in the table, the three containing about 40 per cent of moisture had a fuel value of 6,079 to 6,241 B.t.u., while the Texas lignite with a moisture content of 33 per cent, had a fuel value of 6,840 B.t.u. The percentage of moisture removed in the process of briquetting ranged from 24 to 32 per cent, and the heat value of the briquets was 36.5 to 54 per cent higher than that of the raw lignites.

Excessive moisture in fuel not only causes a waste of useful heat during combustion, because the moisture is vaporized and the vapor superheated, but also is a source of expense to the consumer, who pays freight charges on useless water. For both these reasons lignite briquets have the advantage over raw lignite. In the case of one of the North Dakota lignites the removal of 32 per cent of moisture during briquetting permits a decided lessening of the cost of supplying a consumer with a given number of heat units. The advantage of the briquets in this respect is of special importance when transportation to a distant market is involved. If the briquets possessed no other advantage over raw lignite than their higher heat value, they would be worth 50 per cent more than the raw fuel. The table shows that the average heat value of the different lots of briquets was about 9,300 B.t.u. in each case, although the lignites from which they were made came from three States widely separated, and the heat values of the raw fuel varied considerably.

RESISTANCE TO WEATHERING

The results of the weathering tests indicate that the lignite briquets resist weathering much better than the raw fuel. Some samples of briquets after exposure to autumn and winter weather for four months were in nearly as good condition as at first. Samples stored under cover did not show any signs of deterioration after six months, although the raw lignite stored under the same conditions turned to slack in two months.

RESISTANCE TO HANDLING

The briquets bear handling better than the raw lignite. Their superiority is, of course, more striking after the lignite and the briquets made from it have been stored for several weeks.

EFFICIENCY AS FUEL

Preliminary results of producer gas and steaming tests indicate that as gas-producer fuel the briquets were 43 per cent more efficient and as boiler fuel were 14.5 per cent more efficient than the raw lignite.

WORK OF NORTH DAKOTA EXPERIMENT STATION

The State of North Dakota has lately interested itself in the development of its lignite resources to the extent of passing laws requiring all its public schools to use lignite for heating wherever possible.

In that State also an experimental plant has been established at Hebron by act of legislature as a sub-station of the mining engineering department of the University of North Dakota to "foster the development of mineral and allied industries by providing for experimentation, encouragement, publicity, and practical tests," etc., and the investigations have been started by Prof. E. J. Babcock of the gas making and briquetting qualities of the lignites of the State. Briefly the method he has developed is to distil the raw lignite in retorts, saving the gas, tar and ammonia and briquetting the residue from the retort with a composite binder of coal-tar pitch and a starchy material. In a letter of recent date he states that at the present time there is no commercial briquetting plant in the State of North Dakota, although there have been numerous promotion schemes projected from time to time within the State, but these in nearly if not every case have been purely paper promotion projects. They have had no legitimate basis for their existence and have never even gone so far as to really attempt earnestly to establish a plant. Further he states that none of the various processes which have been hitherto suggested for the treatment of North Dakota lignite have been adapted to the needs of a successful briquetting industry. As a result of his research he has come to believe that North Dakota lignite could not be made of very great commercial value except in the immediate vicinity of the mines unless it could be raised to a higher fuel value so that it would compare more nearly with other standard coals, and so that it would stand long exposure to the weather without slacking and would stand up in the fire and burn also without slacking. To accomplish this purpose, he has developed a method of removing and saving the gas and other by-products and briquetting the residue, which has a high fuel value but which does not coke, and would, therefore, be worthless unless briquetted. He also states that this residue, however, when briquetted becomes a splendid fuel, nearly equal in heat value to the average anthracite, and these briquets stand the weather and retain their form almost perfectly while burning. By this method by-products and the gas are saved and the gas has excellent heating and lighting properties when used with the mantle. Also his type of briquet is strong, stands weathering and shipping well, and is of such high heat value that the product can be transported to a considerable distance and marketed at a profit.

In his bulletin it is stated that the gas obtained has a heat value of about 400 heat units per cubic foot and the briquets made from the residue average in heat value about 12,000 to 12,500 heat units, or about the same as anthracite coal.

COST OF BRIQUETTING LIGNITE

The cost of briquetting lignite should be considered under two heads; namely,

(*a*) Cost of briquetting without binder by the German method.

(*b*) Cost of briquetting with binder on low pressure machines.

COST OF PRODUCTION

For One Two-Press Plant Capacity 120 Long Tons of Briquets in 24 Hours or 3000 Tons per Month

Item			Monthly	Per ton of Briquets
1. Interest and depreciation:				
(*a*) Interest 5% on $111,000			$462.00	
(*b*) Depreciation: 10% of $66,000, machinery and equipment			550.00	$0.400
5% of $45,000, building			188.00	
2. Raw Lignite:				
(*a*) Lignite for briquetting (for 1 ton briquets 68 bu. at 1.05 cents per bu.)				0.712
(*b*) Lignite for boiler fuel, (for 1 ton briquets 34 bu. at 1.05 cents per bu.)				
Total—102 bu. at 1.05 cents or monthly 3000 x 102 x 1.05 cents			3215.00	
3. Labor for 24 hours (2 shifts):				
2 engineers at	83.3 cts.	$1.66		
2 firemen	83.3 cts.	1.66		
1 ash remover	71.4 cts.	.71		
3 coal passers	83.3 cts.	2.50		
8 drier fillers	83.3 cts.	6.66		
2 pressmen	83.3 cts.	1.66		
1 machinist	83.3 cts.	.63		
12 boys for loading and piling	53.3 cts.	6.40		
31 men	Total	22.08 per day x 25	552.00	.231
2 foremen 1 weighman	at $47.60 per month		142.80	
34 men total				
4. Oil, waste, and lighting material			176.00	.059
5. Repairs, maintenance, etc.			125.00	.042
Total				$1.800

The cost of briquetting without binder by the German method will of course vary to some extent with the locality where the plant is located. The fact that lignite is high in moisture makes it even more imperative that a plant for briquetting lignite should be located at the mine than in the case of a plant for briquetting bituminous or anthracite coals, as latter type of plants may frequently be located to better advantage away from the mines near the point of consumption of the briquetted fuel.

In considering the cost of briquetting lignite without binder, it will be best to first study actual figures obtained in a plant operated in Germany.

In Germany from one and one-half to two tons of raw lignite are required to make one ton of briquets.

Mr. G. Franke in his "Handbuch der Brikettbereitung" gives figures (from which the foregoing was translated), on the cost of briquetting lignite in a German plant having two presses and a capacity of 120 long tons per day of 24 hours. The German weights and costs have been converted into American units for easier comparison.

Hence the briquetting cost is approximately $1.80 per ton of briquets in Germany.

The main item in the cost of briquets in Germany is the cost of the raw lignite and amounts to from 55 to 70 per cent of the total cost according to a German authority.

Considering now the equivalent briquetting cost in the United States, using an imported German machine and the equipment necessary, all imported, there will have to be added to the German cost of $66,000 a duty of 45 per cent under the present tariff which will amount to $29,700 or approximately $30,000. The total cost of the machinery would then be $96,000.

The cost of building would no doubt be less here than in Germany, as it is not customary to provide as elaborate a building here for industrial purposes as it is in Germany. Again probably much of the auxiliary equipment to the press could be bought in the United States at, at least, as low a cost as in Germany. On the other hand, more conveyors and elevators would be found economical in this country where labor is higher, than abroad.

The cost of a two press briquetting plant, equipped with German presses and driers but in which American auxiliary equipment is used and all housed in an American style of mill building of steel and concrete walls, should not exceed $100,000. Such a plant would have a capacity of about 100 tons of briquets per day of 20 hours or 2500 tons per month of 25 working days. Then the cost of production in America would be somewhat as follows:

COST OF PRODUCTION IN UNITED STATES.

PLANT WITH TWO GERMAN PRESSES AND AMERICAN EQUIPMENT; CAPACITY OF PLANT 100 TONS PER 20 HOURS OR 2500 TONS PER MONTH OF 25 WORKING DAYS

			Monthly	Per ton of Briquets
1. Interest and depreciation:				
(*a*) Interest 5% on $100,000			$417.00	$0.458
(*b*) Depreciation: 10% on $75,000, machinery and equipment			625.00	
5% on $25,000, buildings			104.00	
2. Raw Lignite:				
(*a*) For briquetting 1.5 tons for 1 ton briquets at $1.50 per ton				2.25
(*b* Lignite for boiler fuel 0.5 ton for each ton briquets at $1.50 per ton				.75
Total, 2 tons at $1.50, or monthly, 3000 x 2 x $1.50			9000.00	
3. Labor for 20 hours (2 shifts)				
2 engineers–pressmen at	$3.00	$6.00		
2 firemen at	1.75	3.50		
2 coal unloaders at	1.75	3.50		
2 briquet loaders at	1.75	3.50		
2 roustabouts at	1.75	3.50		
1 machinist at	3.00	3.00		
11 men — total pay per day		$23.00		
23 x 25		575.00		
1 clerk or weighman at $50 per month		50.00	625.00	0.250
4. Oil waste and light			100.00	.04
5. Repairs, maintenance			125.00	0.05
Total				$3.878

The cost of briquets made in the United States of lignite costing $1.50 per ton at the mines would be about $3.88 per ton. Of this figure the cost of the raw lignite for making the briquet and for boiler fuel at the briquetting plant would be about $3, leaving a briquetting cost exclusive of the cost of raw fuel of 88 cents, or the cost of the raw fuel was 77 per cent of the total cost.

Using the figures given in the first part of this article under the cost of lignite at the mines in 1910, we find that in Texas where the price of lignite at the mine averaged 87 cents per ton, the cost of briquets (if Texas lignite can be briquetted by this process) would be:

2 tons lignite at 87 cents	$1.74
Briquetting costs	.88
Total	$2.62

In North Dakota likewise the cost of briquet per ton would be:

2 tons lignite at $1.49	$2.98
Briquetting costs	.88
Total	$3.86

In California also the cost of briquets per ton would be:

2 tons lignite at $1.64	$3.28
Briquetting costs	.88
Total	$4.16

As the above figures are based on the value of the lignite at the mine, the cost of the lignite and therefore the cost of briquets would be somewhat less if the briquetting was done by the operator of the mine as the raw material used might include slacked lignite which would generally be considered valueless at a lignite mine. However, the above figures are conservative.

Estimates made two years ago in a bulletin of the Bureau of Mines of the cost of briquets made in a one-press plant with lignite at the price in 1907 were per ton of briquets as follows:

For Texas	$2.51
For North Dakota	3.53
For California	5.24

The variation between these figures and those given in the estimate above is due (*a*) to difference in size of plant for what they were figured, and (*b*) difference in prices of fuel.

From a comparison of the cost of briquets made in Germany with the cost as made in the United States, it will be noticed that the briquetting cost in Germany exclusive of the raw fuel used is approximately 73 cents per ton of briquets and in the United States it is 88 cents per ton. This increased price in the United States is due to the increased cost of labor in the latter country and the shorter working hours (ten instead of twelve in Germany) and would be higher still if it were not for the fact that less labor would be required in the United States, on account of the greater efficiency per man and the use of mechanical handling of materials.

Again it will be noticed that the cost of raw lignite to produce one ton of briquets costs in Germany $1.07, while in the United States the equivalent amount costs $3 or nearly three times as much.

Location	Kind of briquet	Size or shape	Weight	Price	
		(Inches)		Wholesale	Retail
Dresden, Saxony	Brown coal			$1.19-1.178 M	$.15-.20 per hund.
	Bituminous	5 x 3 x 5	1.5-1.6 lb	3.68-8.80 M	.38-.77 per hund.
Basel, Switz.	Anthracite				$9.10 per ton
	Brown coal				7.50 per ton
Hamburg, Germany	Bituminous		About 6 lbs.	$4.87 per ton	
	Saxon br. coal		About 6½ lbs	4.04 per ton	
Glasgow, Scotland	Bituminous(?)	6 x 4 x 3½		3.28 to 3.52 per ton	4.25-4.38 per ton
Roubaix, France	Bituminous(?)	Cubes			7.33 per ton
		Large egg			7.33 per ton
		Small egg			7.72 per ton
Havre, France	Bituminous	Large		5.40 per ton	7.53 per ton
		Perforated		5.60 per ton	7.91 per ton
Paris, France	Bituminous	Egg			9.65 per ton
		Perforated			10.80 per ton
		Solid			9.26 per ton
Frankfort, Germany	Anthracite	12 x 8 x 6			4.60 per ton
	Anthracite	Egg			5.00 per ton
	Brown coal with 5% coa tar and asphalt earth	7 x 5 x 2	1 lb.	2.90 per ton	3.25 per ton
	Bituminous	12 x 8 x 6	About 10 lbs.	4.00 per ton	4.40 per ton
	Mixture of bituminous coal, brown coal and tar			2.93 per ton	
Strettin, Germany	Brown coal	6 x 2 x 1	1.3 lbs.	4.05 per ton	5.25 per ton
	Bituminous	6 x 4 x 2½	About 3 lbs.	4.52 per ton	5.75 per ton
Kehl, Germany	Brown coal			3.85 per ton	
	Bituminous			4.55 per ton	5.27 per ton
Rotterdam, Holland	Charcoal			6.83 per ton	
	Bituminous			5.63 per ton	
	Brown coal			4 82 per to	5.43 per ton

A consular report states that in 1908 the average cost of the lignite used for briquetting in Germany was 44 cents per ton, and that the average amount of lignite used per ton of briquets was 2.12 tons; also the average value of the briquets made by the dry process was $2.19 per ton of briquets. At 44 cents a ton the 2.12 tons of lignite used would cost a little over 93 cents. The difference between this figure, 93 cents, and the value of a ton of briquets, $2.19, or $1.26, would be the average cost of briquetting lignite in Germany on a large commercial scale.

In studying the practicability of lignite briquets for American markets, it is interesting to note the prices at which they are sold in Europe, and therefore the foregoing table is presented, the data of which was obtained from Consular reports of 1907. For comparative purposes the price of other types of briquets is also included.

ESTIMATE OF THE COST OF BRIQUETTING AMERICAN LIGNITE WITH BINDING MATERIAL

The cost of briquetting lignite in the United States, using a binding material, will of course vary according to the kind of binding material used and the locality in which the briquetting is done. For comparison of this method of briquetting lignite with that given in the previous estimate where no binder was used, the following estimate may be of interest:

			Per month	Per ton briquets
1. Fixed charges:				
(*a*) Interest on investment 6% on $75,000			$375.00	$0.179
10% on $50,000 (machinery)			417.00	
(*b*) Depreciation 5% on $25,000 (buildings, etc.)			104.00	
2. Labor:				
2 engineer-pressmen at	$3.00	$6.00		
2 firemen at	2.00	4.00		
2 binder-men at	2.00	4.00		
6 laborers at	1.75	10.50		
12 men Total		$24.50		
$24.50 x 25			612.00	0.133
1 clerk or weigher at	$50.00		50.00	
3. Oil, waste and light			50.00	0.010
4. Repairs, maintenance, etc			100.00	0.020
				0.342
5. Binder 6% at $12 of 5000 tons			3600.00	.720
Total briquetting cost				$1.062

Assumption:

Capacity of plant 200 tons per day of 20 hours.
Capacity of plant 5000 tons per month of 25 days.
Capacity of plant 60,000 tons per year.
Plant operated 2 shifts of 10 hours each per day.
Cost of plant:

Machinery	$50,000.00
Building and tracks	25,000.00
	$75,000.00
Cost of binder	$12.00 per ton

COST OF LIGNITE BRIQUETS USING BINDER

The cost of briquetting lignite using a binder, would, from above estimate, be approximately as follows in the States mentioned below:

Locality	Value of raw fuel per ton	Tons of raw lignite per ton briquets	Total cost of raw fuel	Briquetting cost	Cost of briquets per ton at plant
Texas..........	$0.87	2	$1.74	$1.06	$2.80
North Dakota...	1.49	2	2.98	1.06	4.04
California.......	1.64	2	3.28	1.06	4.34

SUMMARY

Investigations have shown therefore that certain classes of lignites can be briquetted without binder, and in that form are much superior to the raw fuel from which they were made, although not equal in fuel value to the higher grades of bituminous and anthracite coals. Also lignite briquets made without binder do not hold together in the fire as well as those made with a binder, nor do they resist the effects of moisture as well.

On the other hand, briquets made without binder supply to a large extent the household fuel in Germany, and so must be considered a practical success there. Also briquets so made resist weathering much better than the raw lignite, have higher heat value, are uniform in size, permitting regular firing, and can be stored for a considerable time without serious deterioration. Again a plant making lignite into briquets without binder is not dependent on a supply of binding material and this is a very important consideration for a plant located in an out-of-the-way district to which binder would have to be brought from a long distance.

It must be remembered, however, that all lignites are not suitable for briquetting without binder and therefore those requiring a binder to agglomerate them are not considered in the above comparison of the relative merits of briquets made with and without binder.

Unquestionably, briquetting improves the fuel value of any lignite and makes it possible for it to compete with bituminous and anthracite coals as a fuel for household use in districts adjacent to lignite deposits.

It requires further investigation to determine whether Texas lignite can be briquetted commercially without binder or not, but results so far obtained by the Bureau of Mines indicate that binder will be required to make entirely satisfactory briquets of lignite from that locality.

North Dakota and California lignites, on the other hand, possess excellent briquetting qualities and probably can be briquetted without binder if it is found desirable to utilize them in this way, that is, if a market be developed for this type of briquet.

The North Dakota and California lignites can probably be made into better commercial briquets to meet American conditions, by the addition of binder which will permit of different shaped briquets being made, which will stand up in the fire better and withstand the effects of weather better than those made by the German binderless process.

There is without question lignite in other localities than Texas, North Dakota and California which will equal them in briquetting qualities, but at present time no extensive investigations have been made of other lignites than those mentioned above.

With fuels at their present prices in North Dakota, it appears feasible for lignite briquets made in that State to compete with other fuels used there, but it is doubtful if the same may be said of the Texas and California fields where fuel-oil and high-grade fuel in adjacent States make the outlook for the commercial success of briquetting less roseate.

UEBER DIE BILDUNG DES ASPHALTES IN ERDOLEN

VON R. ZALOZIECKI UND C. ZIELINSKI
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Vor Jahren hat einer von uns Zaloziecki bei der Einwirkung von Schwefelsäure verschiedenen Grades direkt auf rohes Erdöl eine Bildung des Asphaltes, die auf Polymerisations—und Kondensationser—scheinungen züruckzuführen ist, beobachtet. Es lag nahe zu untersuchen, ob nicht auch andere Körper mit kondensier baren Eigenschaften dieselben Wirkungen ausüben könnten. In der Tat hat sich diese Voraussetzung bestätigt, und es hat sich gezeigt, dass man Asphalt, oder richtiger gesagt asphaltartige Körper auf diesem Wege aus dem Erdöl erzeugen kann und dass mineralische Substanzen dem Erdöl gegenüber asphaltbildende Eigenschaften besitzen. Wir haben in dem Sinne eine Reihe von Kondensationsmitteln unter verschiedenen Bedingungen geprüft und hiebei nachfolgende Resultate erzielt.

Der Untersuchung wurde das Rohöl aus Wankowa-Brelikow, einer Grube in Zentralgalizien unterworfen. Das Rohöl hatte ein spec. Gew. von 0.8539 bei 15° C. einen Paraffingehalt von 1.54%, die elementare Zusammensetzung:

Kohlenstoff	85.29%
Wasserstoff	14.41%
Schwefel	0.19%
Sauerstoff (Diff)	0.11%

und was die Hauptsache ist, es enthielt keinen Asphalt, denn die Alkohol-Aetherprobe ergab ein vollständig negatives Resultat.

Dieses Rohöl wurde in solchem oder in entbenziniertem Zustande mit gepulverten kondensierbaren Substanzen, gewöhnlich 5% auf 20 gr. Rohöl zusammengebracht und der Einwirkung überlassen a. unter Lichtabschluss; b. unter Lichteinfluss beide Male in gewöhnlicher Temperatur; c. unter Erwärmung auf dem Wasserbade und d. durch Erhitzung im geschlossenen Rohr.

Wo es anging wurde öfters geschüttelt oder gemischt. Nach vollzogener Einwirkung wurde der Inhalt der Gefässe stark mit Petroleumbenzin vom spec. Gew. ungefähr 0.7000 verdünnt, filtrirt und das anhaengende Rohöl so lange von Gefäss und den Niederschlag auf dem Filter gewaschen bis das Waschbenzin ganz rein ablief. Der Niederschlag auf dem Filter und die anhängenden Teile an dem Gefässe wurden im warmen Benzol aufgenommen und von dem Ungelösten filtriert, welche Operation so lange wiederholt wurde, bis noch nennenswerte Mengen von Benzol gelöst wurden; in einigen Fällen wurde nachträglich noch mit Schwefelkohlenstoff und hierauf mit Chloroform das in Benzol Unlösliche gelöst. — Das in einem gewogenen Kolben aufgefangene Benzolfiltrat wurde zuerst auf dem Wasserbade vom Benzol abdestilliert und hierauf bei einer Temperatur von 105° C. so lange erwärmt, bis der Benzolgeruch vollständig verschwunden war. Als kondensierende Substanzen wurden der Reihe nach: Na, Na_2S, Na_2CO_3, C_2H_5ONa, S, BaS, BaO_2, P, P_2O_5, Cu_2Cl_2, C_oCl_2, $ZnCl_2$, $FeCl_3$, $AlCl_3$ und MnO_2 verwendet.

In einigen Fällen hat sich unter Einwirkung dieser Substanzen Asphalt aus dem Rohöl und zwar in verschiedenen Mengen gebildet, in anderen erwiesen sich die Substanzen wirkungslos.

I. Versuchsserie: Einwirkung unter Lichtabschluss durch 4 Wochen bei gewöhnlicher Temparatur:

1. mit metallischem Natrium ohne Wirkung.

2. mit Na_2CO_3 ohne Wirkung.

3. mit wasserfreiem Fe Cl_3 erhalten 0.87% eines weichen, bereits in der Kälte in Benzol lôslichen Asphaltes,

4. mit C_2H_5ONa erhalten 0.28% eines gleichfalls weichen leicht in der Kälte benzollöslichen Asphaltes.

5. mit wasserfreiem $AlCl_3$ erhalten 1.28% eines in Benzol schwieriger löslichen, teilweise in Schwefelkohlenstoff und teilweise in Chloroform löslichen Asphaltes,

6. mit wasserfreiem $ZnCl_2$ erhalten 0.63% eines gleichfalls schwieriger löslichen Asphaltes.

7. mit wasserfreiem P_2O_5 erhalten 4.44% eines benzollöslichen Asphaltes,

8. mit wasserfreiem Cu_2Cl_2 erhalten 0.86% Asphalt wie 3.

II. Versuchsserie Einwirkung im Sonnenlichte (im Sommer an der Sonnenseite am Fenster) während 80-tägiger Einwirkungs dauer:

1.	mit C_2H_5ONa	0.322% Asphalt
2.	mit $ZnCl_2$	3.06% Asphalt
3.	mit Cu_2Cl_2	1.12% Asphalt
4.	mit $Al.Cl_3$	3.46% Asphalt
5.	mit P_2O_5	3.17% Asphalt
6.	mit $FeCl_3$	2.12% Asphalt
7.	mit MnO_2	——% Asphalt

8. Rohöl allein produzierte enine geringe Menge Asphalt 0.6%.

III. Versuchsserie. Das Rohôl (150 gr.) wurdemit kondensierenden Substanzen auf dem Wasserbade mit aufsteigendem Kühler durch 166 Stunden (während eines ganzen Monates) erwärmt:

1.	mit C_2H_5ONa	0.70% Asphalt
2.	mit $Al.Cl_3$	7.94% Asphalt
3.	mit Cu_2Cl_2	1.24% Asphalt
4.	mit $ZnCl_2$	2.10% Asphalt
5.	mit P_2O_5	5.37% Asphalt
6.	mit $FeCl_3$	1.13% Asphalt

IV. Versuchsreihe. Das Rohöl wurde mit nachfolgenden Substanzen im eingeschlossenen Rohr durch 35 Stunden auf 140° C. erwärmt:

1.	mit $FeCl_3$	0.64% Asphalt
2.	mit Cu_2Cl_2	0.96% Asphalt
3.	mit $ZnCl_2$	2.48% Asphalt
4.	mit P_2O_5	3.80% Asphalt
5.	mit $AlCl_3$	6.28% Asphalt
6.	mit Na_2S	——% Asphalt
7.	mit S	Spuren Asphalt
8.	mit BaS	——% Asphalt
9.	mit BaO_2	——% Asphalt
10.	mit P	——% Asphalt
11.	mit $S + AlCl_3$	6.37% Asphalt
12.	mit $P + AL\ Cl_3$	6.33% Asphalt

ANMERKUNG: Die Resultate im Einschlussrohr haben sich trotz der höheren Versuchstemperatur, aus dem Grunde geringer erwiesen, als der gebildete Asphalt die kondensierenden Substanzen zusammenbackt und in ihrer weiteren Wirkung stört. Man konnte hiebei naturgemäss keine Mischung und Aufrührung besorgen wie bei Versuchen im Kolben.

Aus den Versuchen ergiebt sich, dass einige Substanzen und zwar ganz verschiedenen chemischen Charakters ein ganz hervorragend asphaltbildendes Vermögen besitzen; von den untersuchten Substanzen wiesen das grösste auf: Aluminiumchlorid und Phosphorsäureanhydrid; im Allgemeinen ist das des ersten grösser, doch giebt es Fälle wie bei Lichtabschluss, wo die Wirkung des P_2O_5 (4.44%) jene des Al Cl_3 (1.28%) stark überwiegt. Man könnte fast versucht sein aus diesem Verhalten Schlüsse über die Art und Weise der Einwirkung zu ziehen, wovon jedochspäter die Rede sein wird.

Nachdem jedoch $Al.Cl_3$ sich als das stärskte asphaltbildende Mittel erwiesen hat. wurde eine grössere Menge des Asphaltes mittelst dessen erhalten dadurch, dass wir entbenziniertes Rohöl mit 5% Al Cl_3 durch 60 Stunden auf 150-155° C. mit aufwärts gerichtetem Kühler erhitzt haben. Hiebei wurde die grösste Menge Asphalt und zwar 10.28% gewonnen.

Von diesem Asphalt waren 6·04% löslich in Benzol
Von diesem Asphalt waren 0·14% löslich in CS_2
Von diesem Asphalt waren 4·10% löslich CH Cl_3

Der Schmelzpunkt des benzollöslichen Asphaltes betrug 61-65° C. und des chloroformlöslichen 95-99° C.

Die Elementaranalyse eines Misch-Asphaltes, den seit seiner Darstellung ein 6-7 wöchentlicher Zeitraum trennte, ergab folgende Resultate:

	am 24/4	2/5	4/6
Kohlenstoff	86.957	86.927	86.048
Wasserstoff	9.090	9.050	9.023
Schwefel	0.906		
Asche	0.300		
Sauerstoff	2.747		

Bei den Verbrennungsanalysen sind die Kalenderdaten angeführt aus dem Grunde, weil sich gezeigt hat, dass sich die Zusammensetzung des Asphaltes bei der Aufbewahrung ändert. Ursprünglich hat eine schlechte Uebereinstimmung der Resultate der Verbrennungsanalyse uns sehr viel Schwierigkeiten gemacht. Wir haben eine ganze Reihe von Elementaranalysen der Asphalte vorgenommen und nur in den seltensten Fällen eine Uebereinstimmung der Resultate erzielt, bis wir schliesslich die Ueberzeugung gewonnen haben, dass die Schuld nicht an der Analyse liegt, sondern an der Veränderlichkeit der gewonnenen Produkte, die eine ausgesprochene Tendenz zur Oxydierung an der Luft besitzen. Dadurch haben wir eine sehr wichtige Tatsache entdeckt, die für die Beurteilung der Bildung des Asphaltes in der Natur von eminenter Bedeutung sein kann.

Der Beweis dafür haben wir gewounen durch die Elementaranalyse eines frisch bereiteten Asphaltes und sukcessive Analysen derselben nach Ablauf von gewissen Zeitfolgen.

Die Elementaranalyse eines frischbereiteten benzollöslichen Asphaltes (mittelst $Al\ Cl_3$) ergab folgende Zahlen am:

	3/6	8/6	12/6
Kohlenstoff	88.742	87.530	87.318
Wasserstoff	8.902	8.797	8.751

Diese Aenderung, welche ziemlich rasch im Anfang vor sich geht, kann nicht auf eine Abdunstung des event. zurückgehaltenen restlichen Lösungsmittels (Benzols) zurückgeführt werden, denn in solchem Falle würde zwar der Kohlenstoffgehalt auch sinken, aber dafür der Wasserstoffgehalt (92.3% C. und 7.7% H.) steigen, sondern muss durch eine Oxydation d. h. durch Aufnahme des Sauerstoffes erklärt werden.

Im übrigen ist die Zusammenstezung der Erdölasphalte eine in gewissen Grenzen verschiedene, je nach der Erzeugungsart. Der chloroformlösliche Asphalt wies in frischbereitetem Zustande nachfolgende Zusammensetzung auf:

	I	II	Mittel
Kohlenstoff	85.534	85.609	85.571
Wasserstoff	9.078	8.993	9.036

Auch die Eigenschaften des Asphaltes sind je nach der Gewinnungsart verschieden. Im Allgemein lässt sich sagen, dass die stärker kondensierenden Mittel als Al Cl_3 P_2O_5. Zn Cl_2 unter denselben Bedingungen einen härteren Asphalt produzieren, die schwächeren Mittel dagegen einen weicheren. Ferner hängt diese Eigenschaft zusammen mit der Dauer der Einwirkung, respekt. Wiederhohlung der Einwirkung, so z. B. kann man durch längere Zeitwirkung, bez. neue Anwendung der schwächeren Mittel denselben Asphalt was Eigenschaftern anbelangt produzieren als mit stärkeren Mitteln rascher. Fener lässt sich behaupten, dass die weicheren Asphalte durchaus in Benzol löslich sind, die härteren dagegen nur teilweise, desgleichen nur teiweise in CS_2, sich jedoch in Chloroform verhältnismässig leicht lösen.

Bei der Bildung von Asphalten tritt mitunter ein deutlicher Geruch nach Schwefelwasserstoff auf und scheinen die Schwefelverbindungen auch an der Asphaltbildung teilzunehmen, wie man aus dem Gehalt des Rohöles und des Asphaltes an Schwefel entnehmen kann.

Im Rohöl haben wir 0.19% S, im Asphalt, von dem mit Al Cl_3 über 10% gebildet werden, rund 0.9, daher fünf mal so viel, oder man kann sagen, dass die Hälfte des ganzen Schwefels im Erdöl in den Asphalt übergegangen ist.

Die gewonnenen Asphalte enthalten stets Aschebestandteile, selbst nach öfteren Filtrieren durch dichtes Filtrierpapier bleibt bei der Verbrennung etwas Asche zurück. Ob das mechanisch verunreinigende Asche ist oder aus gebundenen mineralischen Bestandteilen, bleibt unentschieden, jedenfalls bestätigt das die früher öfters gemachte Beobachtungen.

Durch bedeutend grösseren Aschegehalt zeichnet sich besonders der chloroformlösliche Teil des: Al Cl_3 Asphaltes aus und zwar hauptsächlich in frischem Zustande.

Ueberhaupt scheint das Verhalten des Al Cl_3 ein ganz eigentümliches zu sein. Bei der Einwirkung auf Erdöl im Lichte entwickelt sich sehr viel HCl. und findet intensive Asphaltbildung, wobei sich förmlich eine ganz einheitliche Masse aus dem Al Cl_3 und dem gebildeten Asphalte formt. Man könnte versucht sein anzuneemen, dass da eine Doppelverbindung zwischen gewissen, selbstverständlich ungesättigten, Verbindungen der Erdöle event. auch unter Chlorwasserstoffaddierungsvorgängen (im Dunklen ist die Asphaltbildung gering) mit dem Alluminiumchlorid bez. Alluminiumoxychlorid entsteht, welche infolge Kondensation sich weiter zersetzt. Für diese Art von Vorgängen spricht eine scheinbare Löslichkeit dieser Alluminiumchlorid-Asphaltverbindung in Chloroform, die je auch auf Grund von ganz labilen Anlagerung von Chloroformmolekülen an diese Doppelverbindung vor sich gehen kann. Diese Doppelverbindungen zersetzen sich jedoch sehr rasch, schon beim Filtrieren und in dem Chloroformfiltrat findet man nur noch einen verhältnissmässig kleineren Aschengehalt. Der auf dem Filter verbleibende stark kohlenhaltige Rückstand wies bei der Analyse 7.43% Chlor neben 23.27 % Alluminium auf.

Eine sehr interessante Erscheinung bildet die Beobachtung, dass ein Teil des chloroformlöslichen Asphaltes mit der Zeit benzollöslich wird. Dieser Uebergang vollzieht sich unter Abscheidung von unorganischen Sedimenten und das führte uns hauptsächlich zu den vorhin auseinandergesetzten Annahmen.

Wie weit andere Kondensationsmittel, gleichfalls event.unter ursprünglicher Anlagerung an ungesättigte Bestandteile des Erdöles, auf dasselbe reagieren, lässt sich angesicht der Schwierigkeit der Erfassung solcher Uebergangsstufen nichts sagen, ausgeschlossen ist ein solcher Reaktions-mechanismus keineswegs.

Was sich momentan aus unseren Untersuchungen, die keineswegs noch abgeschlossen sind, sagen lässt, ist, dass sich asphaltähnliche Körper aus dem Erdöl durch Kondensation event. auch Polymerisation mit Hilfe gewisser mineralischer Substanzen bilden und dass diese asphaltähnlichen Substanzen leicht der Oxydation verfallen und in Asphalte ûbergehen. Damit wäre auch der Weg der Asphaltbildung in der Natur aus den Erdölen vorgezeichnet.-Welcher Art Bestandteile es sind, die dieser Asphaltkondensation verfallen lässt sich momentan nicht sagen, ebenso ist die Rolle, die der Schwefel event. Schwefelverbindungen hiebei spielen, nicht klargelegt. Es unterliegt keinen Zweifel, dass sich die Schwefelverbindungen an der Kondensation beteiligen, aber andererseits scheint das Kondensationsvermögen der Erdölbestandteile darauf nicht beschränkt zu sein undes liegt der Annahme der Bildung schwefelfreier Asphalte nichts im Wege.

Unter anderen wird auch das Gegenstand unserer weiteren Untersuchungen bilden.

Date Due

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